LA-UR-21-25702
Approved for public release; distribution is unlimited.

Title: Using vortex dynamics tools to explore magnetic configurations in non-superconducting materials

Author(s): Civale, Leonardo
Nekrashevich, Ivan
Zapf, Vivien

Intended for: Web

Issued: 2021-06-16

Using vortex dynamics tools to explore magnetic configurations in non-superconducting materials

A different look at the magnetic properties of the geometrically frustrated spin-chain compound $Ca_3Co_2O_6$

Leonardo Civale

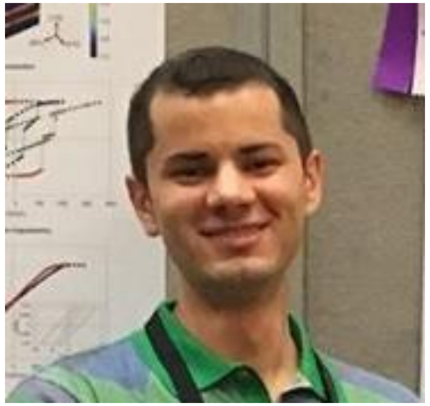
Materials Physics and Applications-Quantum,
Los Alamos National Laboratory

VORTEX 2021

18th International Online Vortex -2021 Conference
May 27th to June 4th, 2021

Research supported by the US DOE, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering.

The team



Ivan Nekrashevich
LANL

Vivien Zapf
LANL

Outline

- Introduction to the magnetic properties of $Ca_3Co_2O_6$
- Ground state and magnetization loops: the puzzle of metastable states that are in the wrong place and relax in the wrong direction
- Magnetic history dependence and how to access the different states
- Using vortex dynamics concepts to try to make sense of the time evolution
- Identification of a previously unobserved stable phase
- The simpler (and vortex-like) dynamics at high T
- Summary

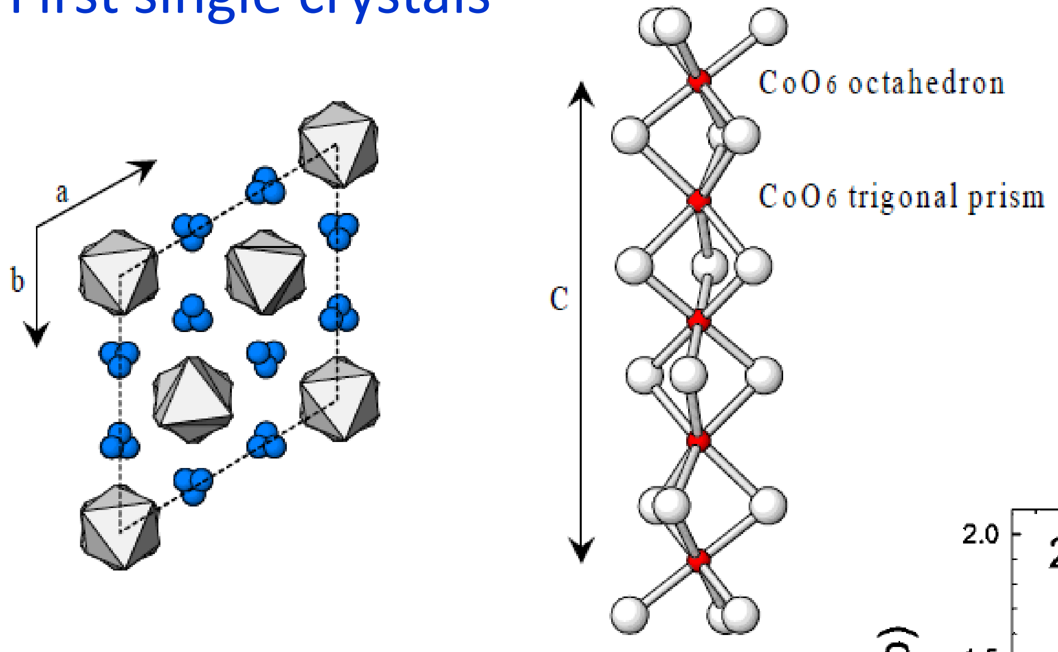
Los Alamos NATIONAL LABORATORY EST.1943

Office of Science U.S. DEPARTMENT OF ENERGY

The geometrically frustrated spin-chain compound $Ca_3Co_2O_6$

A. Maigman et al., Eur. Phys. J. B **15**, 657 (2000)

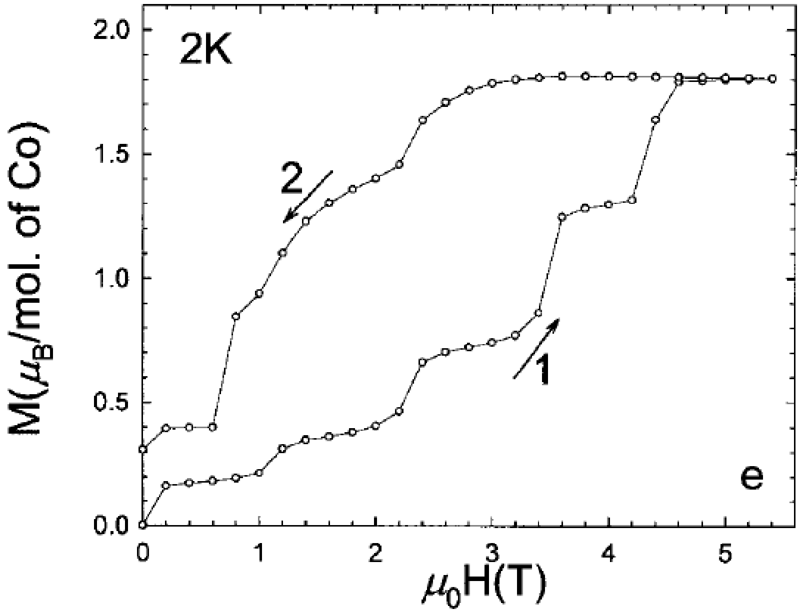
First single crystals

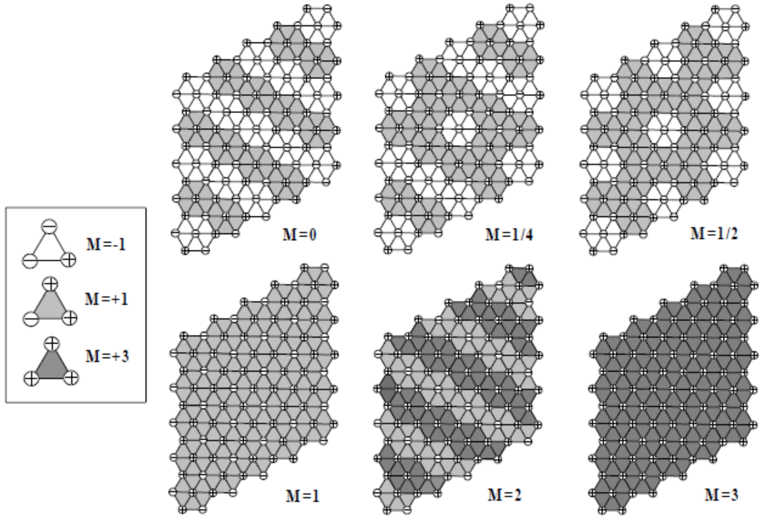


a b

- Rhombohedral structure composed of $[Co_2O_6]_\infty$ infinite chains running along the *c* axis of the hexagonal cell, with the Ca cations located in between them.
- The chains are made of alternating, facesharing CoO_6 trigonal prisms and CoO_6 octahedra.
- Each chain is surrounded by six equally spaced chains forming a triangular lattice in the *ab* plane.
- The intrachain coupling is ferromagnetic (T_{c1}~24K) while the much weaker interchain coupling is antiferromagnetic (T_{c2}~12K).
- The spins in each chain are either all up or all down.

Below 5 K the M(H) loops are irreversible, and reveal new stable magnetic structures …. for *T* = 2 K, two intermediate magnetization plateaus are observed….





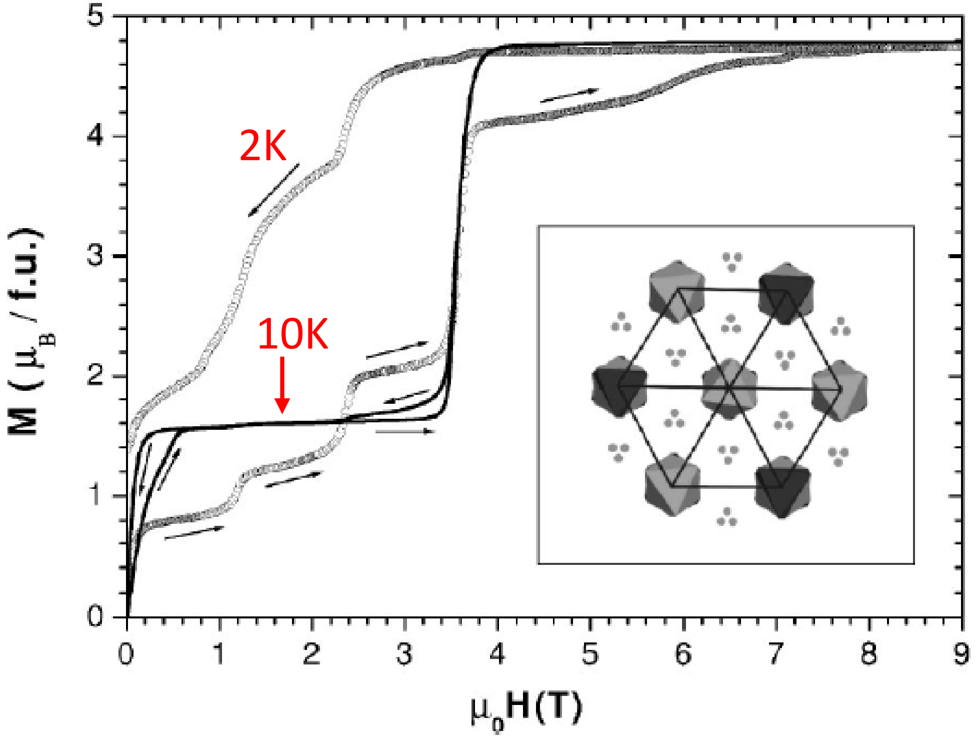
The metastable steps in the $Ca_3Co_2O_6$ magnetization loops

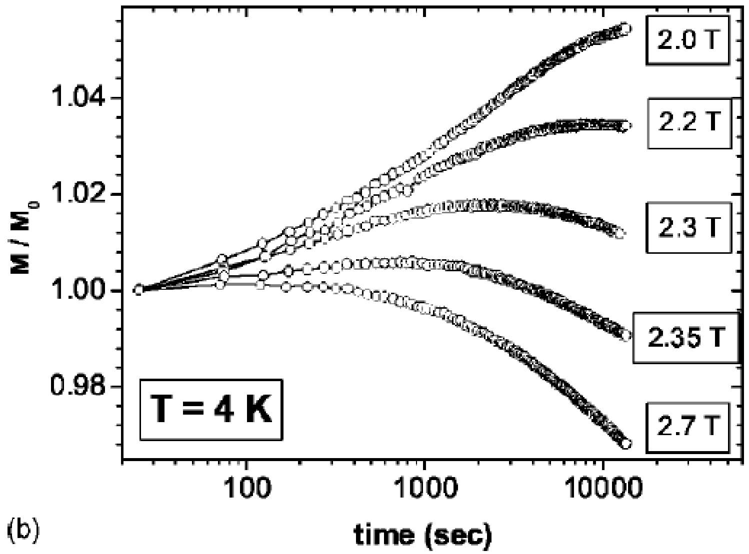
Phys . Rev. B **70**, 064424 (2004)

Temperature and time dependence of the field-driven magnetization steps in $Ca_3Co_2O_6$ single crystals

V. Hardy,[1,2] M. R. Lees,[1] O. A. Petrenko,[1] D. McK. Paul,[1] D. Flahaut,[2] S. Hébert,[2] and A. Maignan[2]
[1]*Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom*
[2]*Laboratoire CRISMAT, UMR 6508, Boulevard du Maréchal Juin, 14050, Caen Cedex, France*
(Received 7 January 2004; revised manuscript received 15 April 2004; published 31 August 2004)

For the spin-chain compound $Ca_3Co_2O_6$, the magnetization curves as a function of the magnetic field are strongly out-of-equilibrium at low temperature, and they exhibit several steps whose origins are still a matter for debate. In the present paper we report on a detailed investigation of the temperature and time dependence of these features. First, it is found that some of the magnetization steps can disappear as the characteristic time of the measurement is increased. A comparison of the influence of temperature and time points to the existence of a thermally activated process that plays an important role in determining the form of the magnetization curves. Second, direct investigations of the magnetic response as a function of time show that this thermally activated process competes with a second relaxation mechanism of a very different nature, which becomes dominant at the lowest temperatures. These results shed new light on the peculiar magnetization process of this geometrically frustrated, Ising-like spin-chain compound.





- Magnetic frustration.
- The time evolution of the metastable states can be *nonmonotonic.*
- The dynamics is very slow (hours to days).

Evolution of the phase fractions studied by neutron diffraction

PRL **106**, 197204 (2011) PHYSICAL REVIEW LETTERS week ending 13 MAY 2011

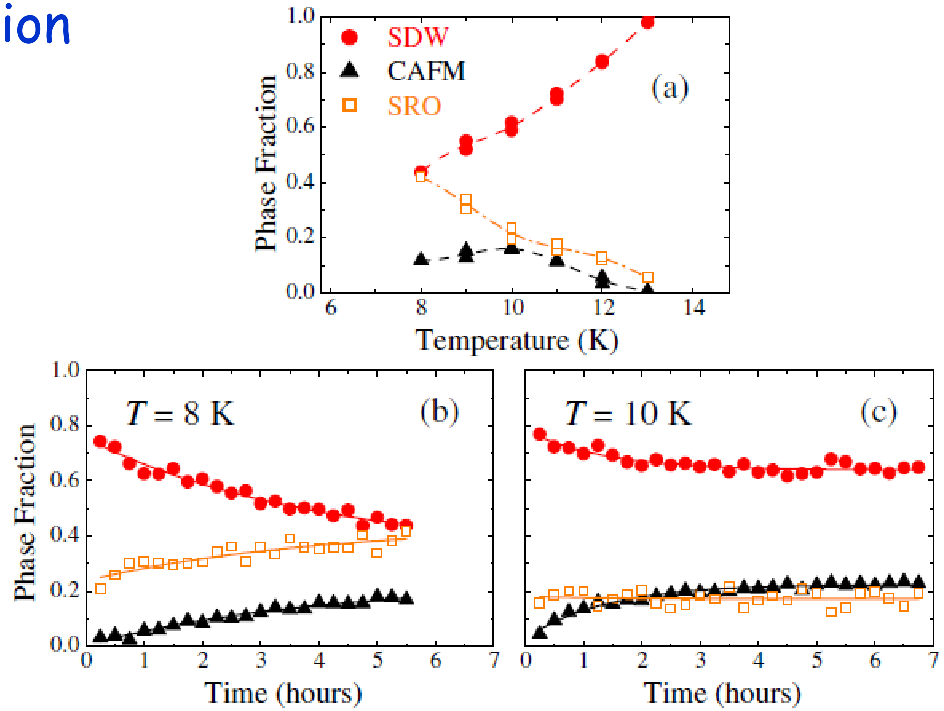
Slow Magnetic Order-Order Transition in the Spin Chain Antiferromagnet $Ca_3Co_2O_6$

S. Agrestini,[1,2] C. L. Fleck,[3] L. C. Chapon,[4] C. Mazzoli,[5] A. Bombardi,[6] M. R. Lees,[3] and O. A. Petrenko[3]

[1]Laboratoire CRISMAT, CNRS UMR 6508, ENSICAEN—Boulevard du Maréchal Juin, 14050 Caen, France
[2]Max Planck Institute for Chemical Physics of Solids, Nöthnitzerstrasse 40, 01187 Dresden, Germany
[3]Department of Physics, University of Warwick, Coventry, CV4 7AL, United Kingdom
[4]ISIS facility, Rutherford Appleton Laboratory, Didcot OX11 0QX, United Kingdom
[5]European Synchrotron Radiation Facility, B.P. 220, 38043 Grenoble Cedex 9, France
[6]Diamond Light Source Ltd., Rutherford Appleton Laboratory, Didcot OX11 0DE, United Kingdom

(Received 15 December 2010; published 13 May 2011; publisher error corrected 17 November 2011)

Using powder neutron diffraction, we have discovered an unusual magnetic order-order transition in the Ising spin chain compound $Ca_3Co_2O_6$. On lowering the temperature, an antiferromagnetic phase with a propagation vector $\mathbf{k} = (0.5, -0.5, 1)$ emerges from a higher temperature spin density wave structure with $\mathbf{k} = (0, 0, 1.01)$. This transition occurs over an unprecedented time scale of several hours and is never complete.



Journal of the Physical Society of Japan **80** (2011) 034701 FULL PAPERS
DOI: 10.1143/JPSJ.80.034701

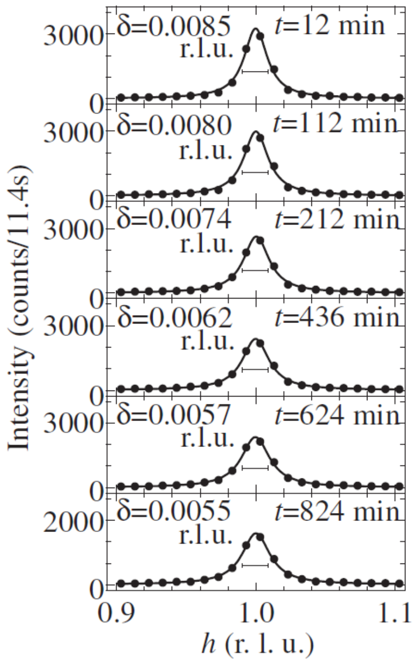
Incommensurate Magnetic Structure and Its Long-Time Variation in a Geometrically Frustrated Magnet $Ca_3Co_2O_6$

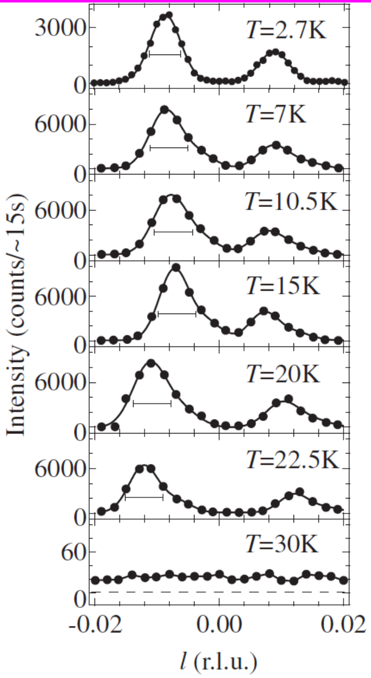
Taketo MOYOSHI* and Kiyoichiro MOTOYA

Department of Physics, Faculty of Science and Technology, Tokyo University of Science, 2641 Yamazaki, Noda, Chiba 278-8510, Japan

(Received September 17, 2010; accepted December 13, 2010; published online February 25, 2011)

Magnetization and neutron scattering measurements have been performed on a single crystal of the magnetically frustrated compound $Ca_3Co_2O_6$. This compound has no random magnetic interactions, but has frustrating magnetic interactions arising from its crystal structure. In this material, a long-time variation in magnetic structure has been observed. After cooling the sample below the transition temperature $T_{c2} = 13$ K, the magnetic Bragg peaks at $(1, 0, \pm\delta)$ corresponding to an incommensurate antiferromagnetic structure move toward (1, 0, 0) with time. The characteristic time for this variation follows the Arrhenius law with an activation energy $E_a/k_B \sim 53$ K. A new magnetic structure model of this material is also presented.





Recent pulsed fields studies at LANL

PHYSICAL REVIEW B **98**, 024407 (2018)

Metastable states in the frustrated triangular compounds $Ca_3Co_{2-x}Mn_xO_6$ and $Ca_3Co_2O_6$

Jae Wook Kim,[1,2] E. D. Mun,[1,3] X. Ding,[1] A. Hansen,[1,4] M. Jaime,[1] N. Harrison,[1] H. T. Yi,[2] Y. Chai,[5] Y. Sun,[5] S. W. Cheong,[2] and V. S. Zapf[1]

[1]*National High Magnetic Field Laboratory (NHMFL), Los Alamos National Laboratory (LANL), Los Alamos, New Mexico 87545, USA*

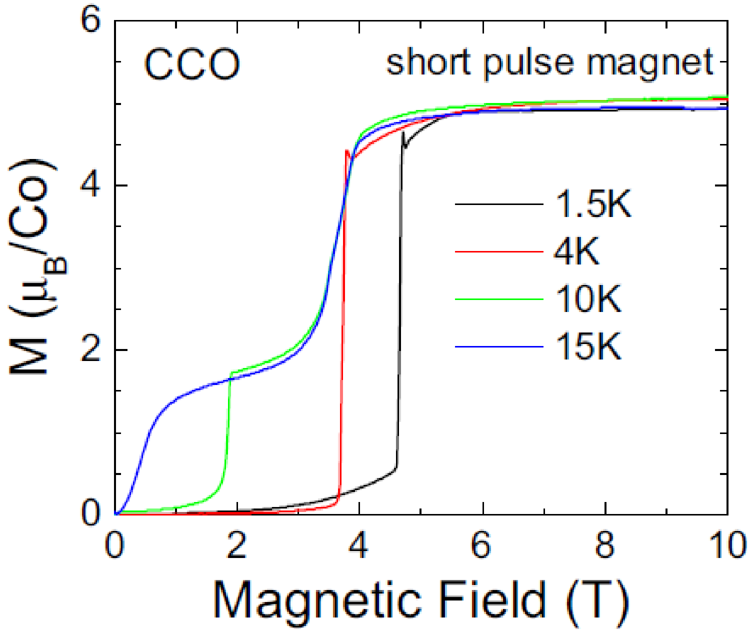
[2]*Rutgers Center for Emergent Materials & Department of Physics and Astronomy, Piscataway, New Jersey 08854, USA*

[3]*Department of Physics, Simon Fraser University, Barnaby, BC, Canada V5A 1S6*

[4]*University of California Los Angeles, Westwood, California, USA*

[5]*Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Bejing 100190, P. R. China*

The observation of unusual metastable behavior in $Ca_3Co_2O_6$ remains an ongoing puzzle. When the magnetic field is increased at certain very slow rates, evenly spaced steps occur in the magnetization and other physical quantities as a function of magnetic field every 1.2 T. The ground state without steps is approached by slow relaxation processes over hours to days. It is a striking example of extremely slow dynamics arising from geometrical frustration in an otherwise clean and long-range ordered system, and its precise description remains controversial. Here we shed light on the mystery by reporting similar behavior in isostructural $Ca_3Co_{2-x}Mn_xO_6$,

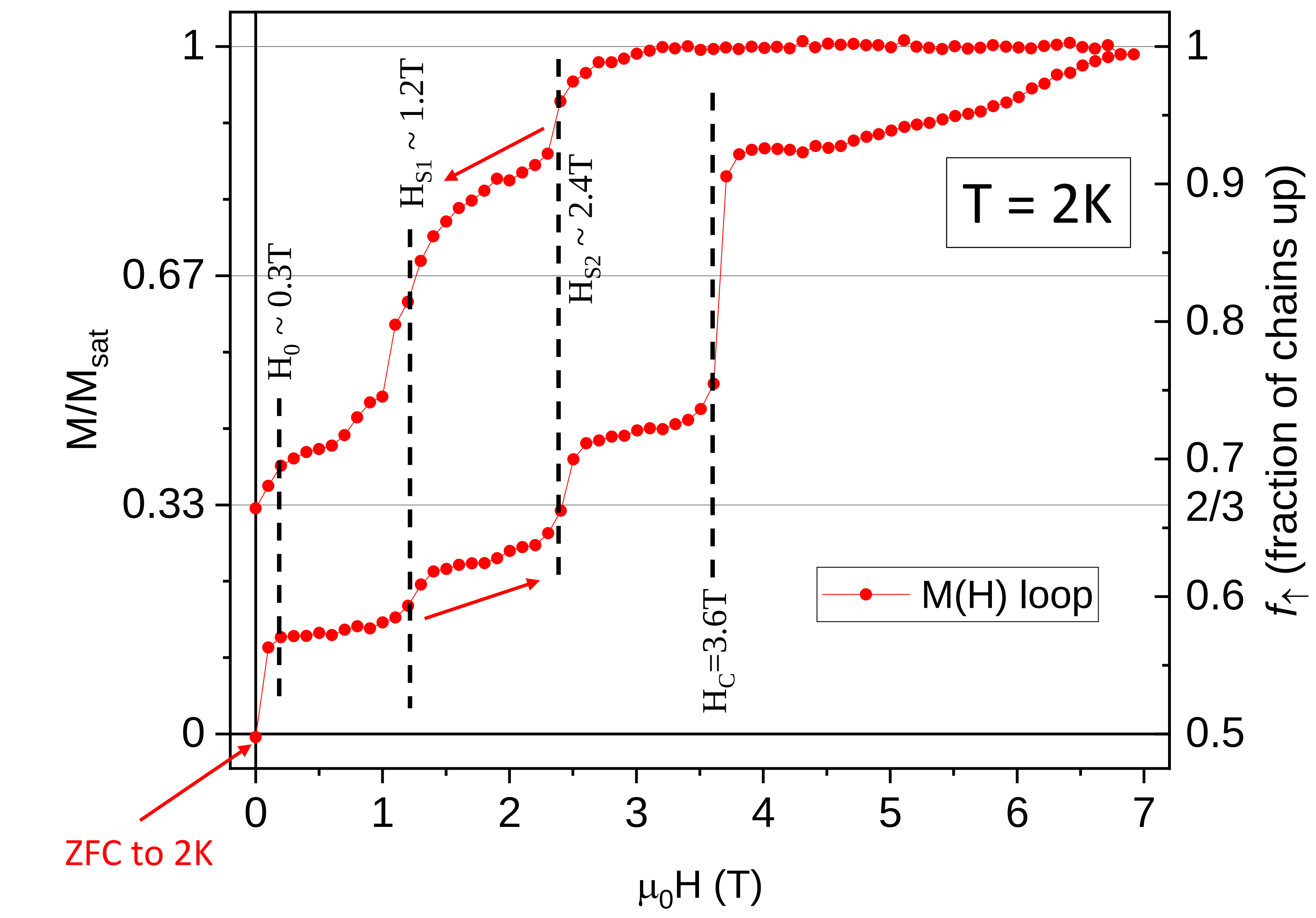


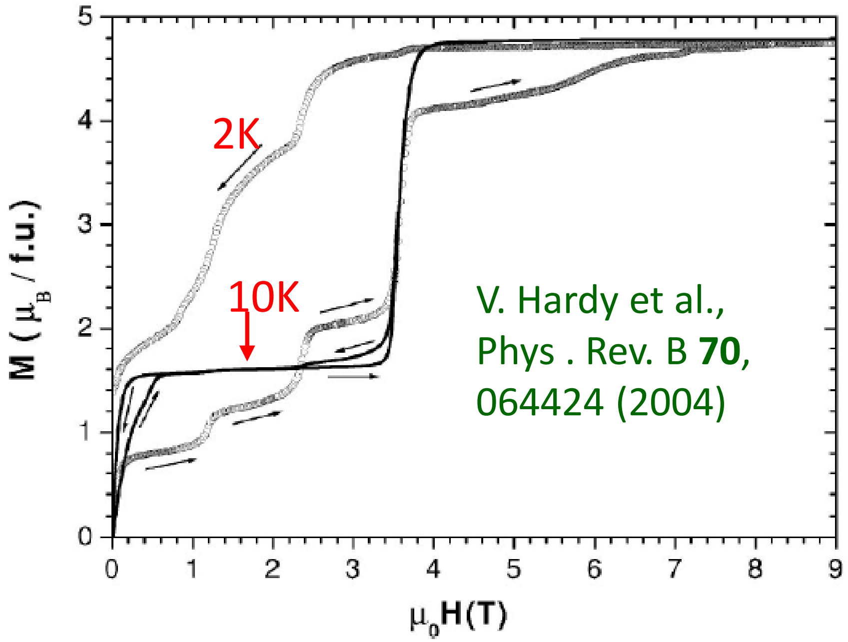
Motivation:
Huge $d\mu_0H/dt$ up to 1660 T/s

Vivien Zapf proposed us to test experimentally some theoretical predictions about quantum annealing
→ Ivan Nekrashevich *et al.*, manuscript in preparation

This talk is about a different story

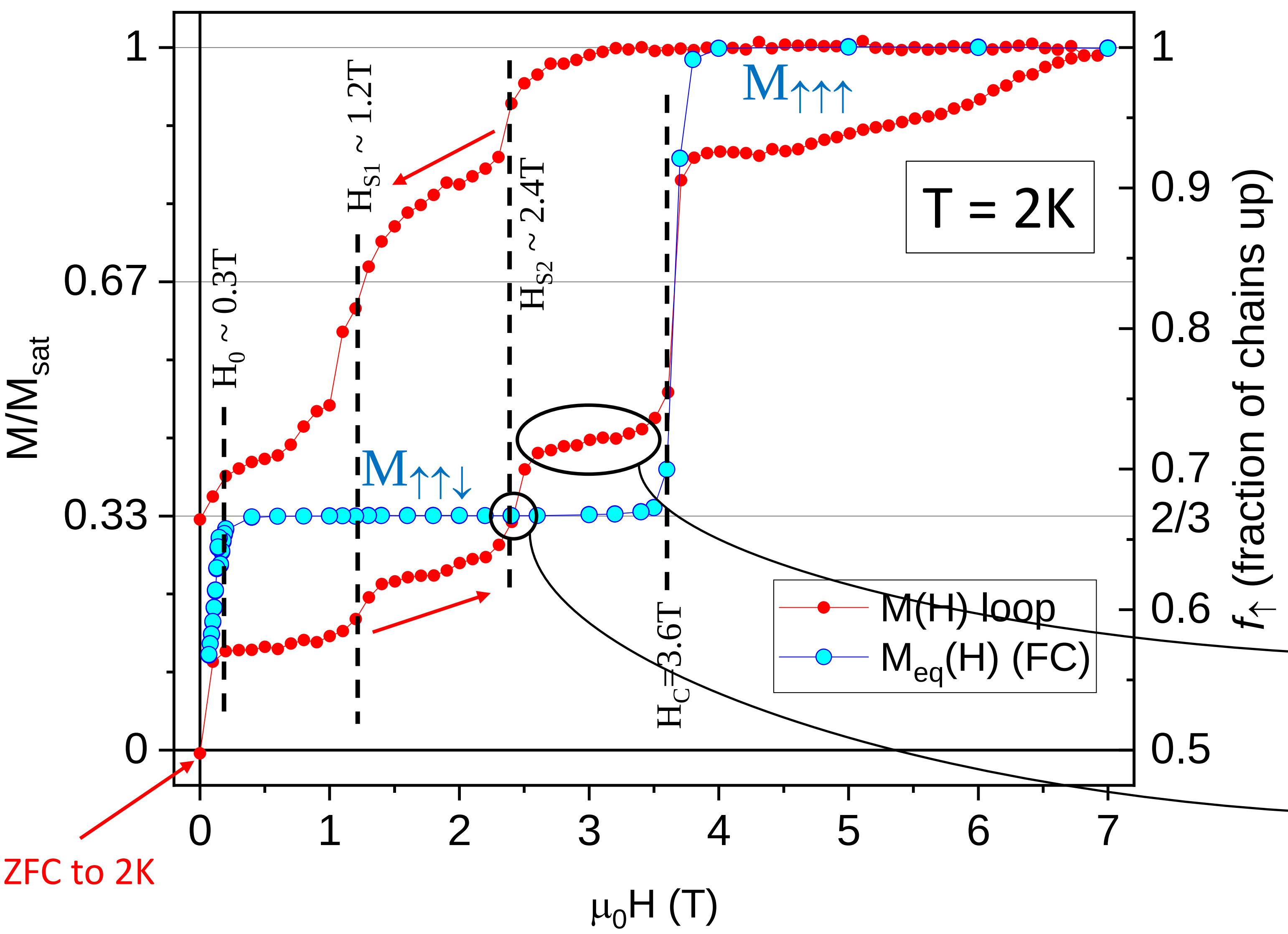
The basics: at T = 2K, we observe the steps at 1.2; 2.4 and 3.6T
Question: which is the ground state at 2K?

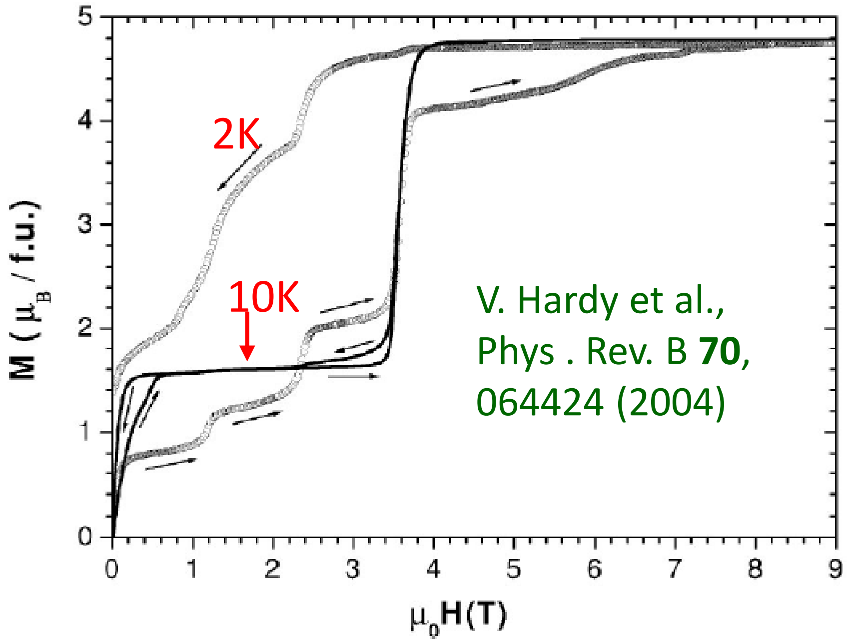




The basics: at T = 2K, we observe the steps at 1.2; 2.4 and 3.6T
Question: which is the ground state at 2K?

Procedure: FC at field H from >30K to 2K – repeat for each H





Ground state:

H>3.6T: all chains up → M_{sat}

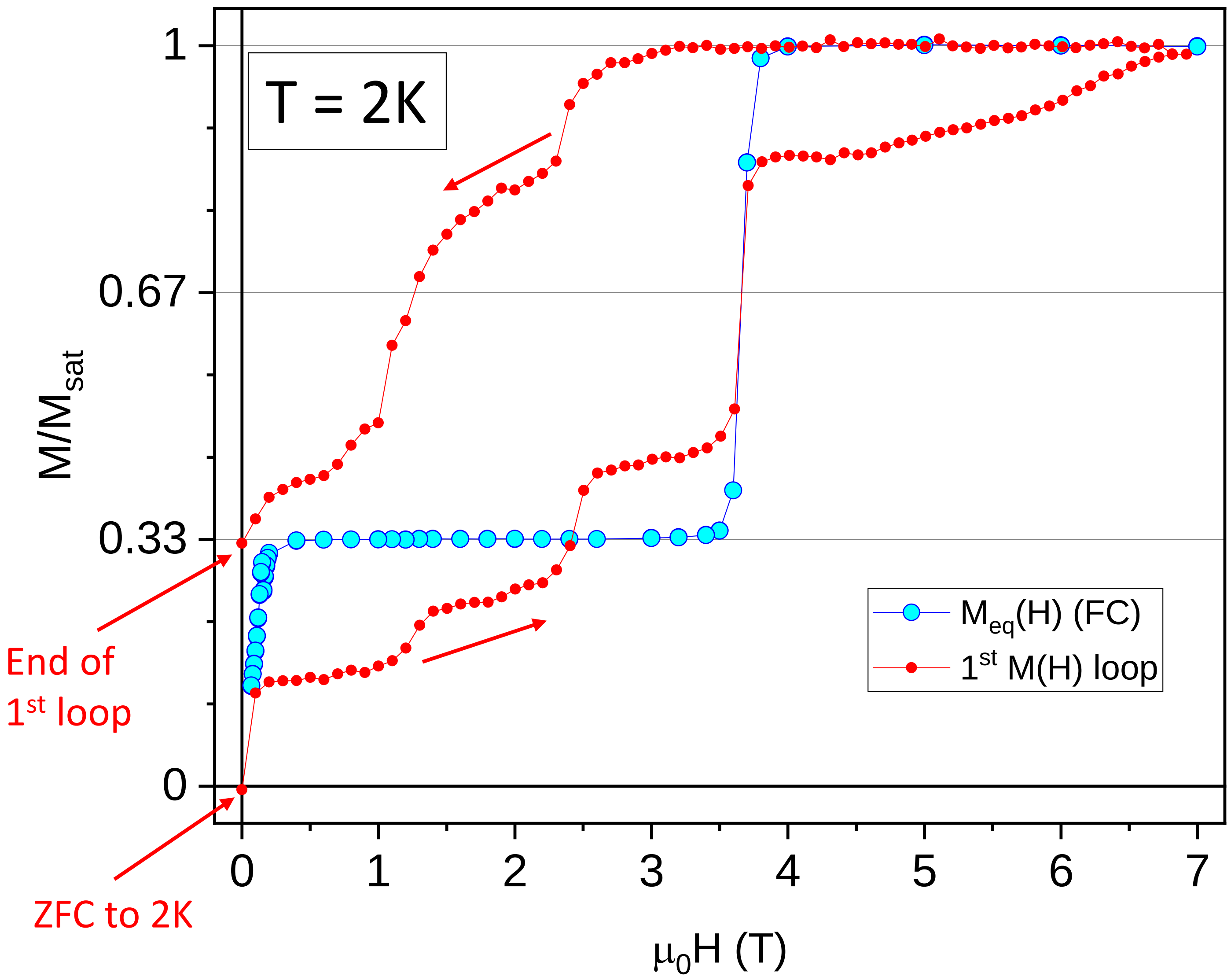
H<3.6T: 2 chains up, 1 down→ $M_{sat}/3$ (Ordered)

Lower branch of the M(H) loop is **above** $M_{eq}(H)$

Curves cross:

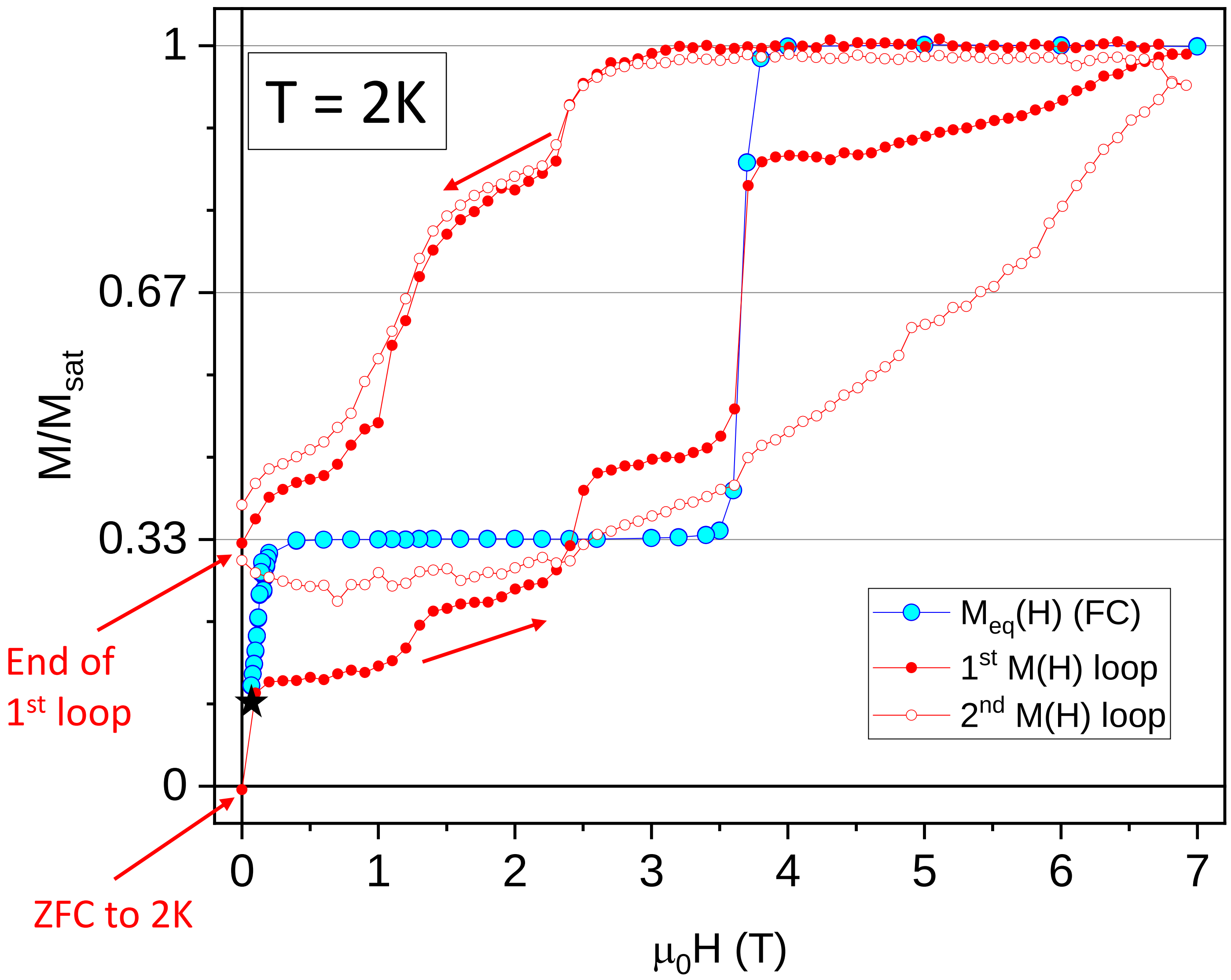
- Same M, different microstate
- Energy barrier

Magnetic history dependence: How do we access the different states?



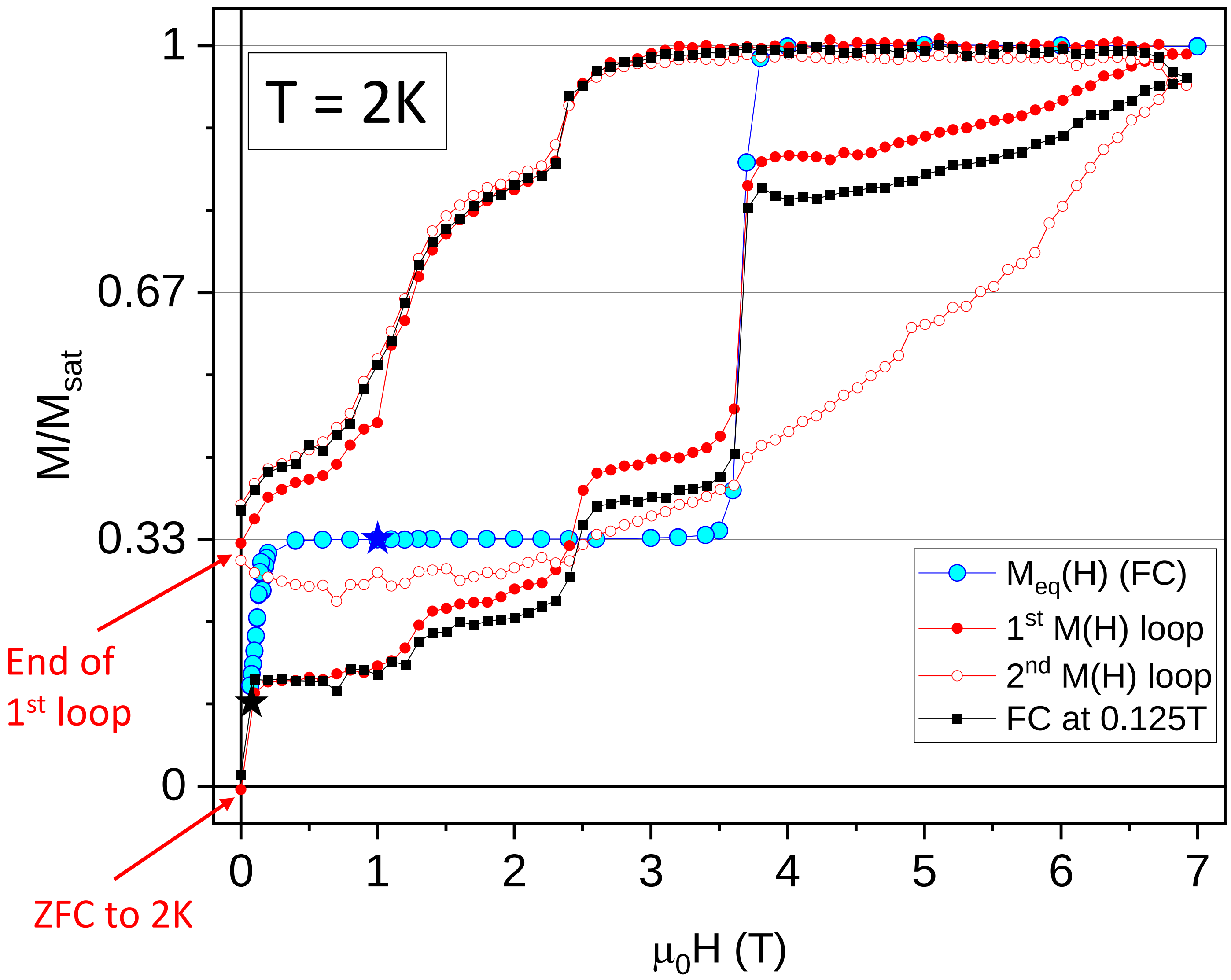
- FC: ordered ground state
- First M(H) loop after ZFC: steps

Magnetic history dependence: How do we access the different states?



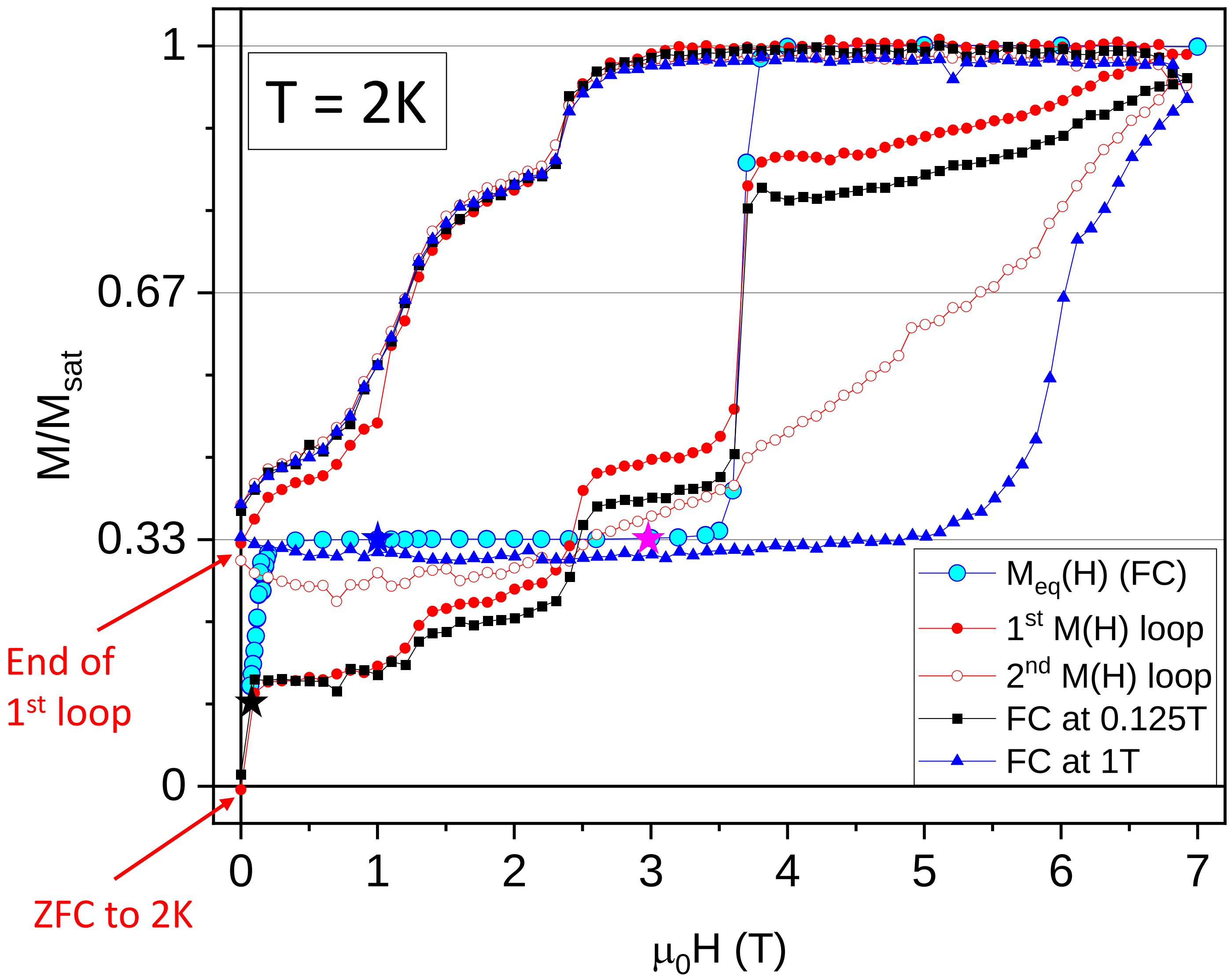
- FC: ordered ground state
- First M(H) loop after ZFC: steps
- Subsequent loops: no steps
- FC at 0.125T → H=0 → loop:

Magnetic history dependence: How do we access the different states?



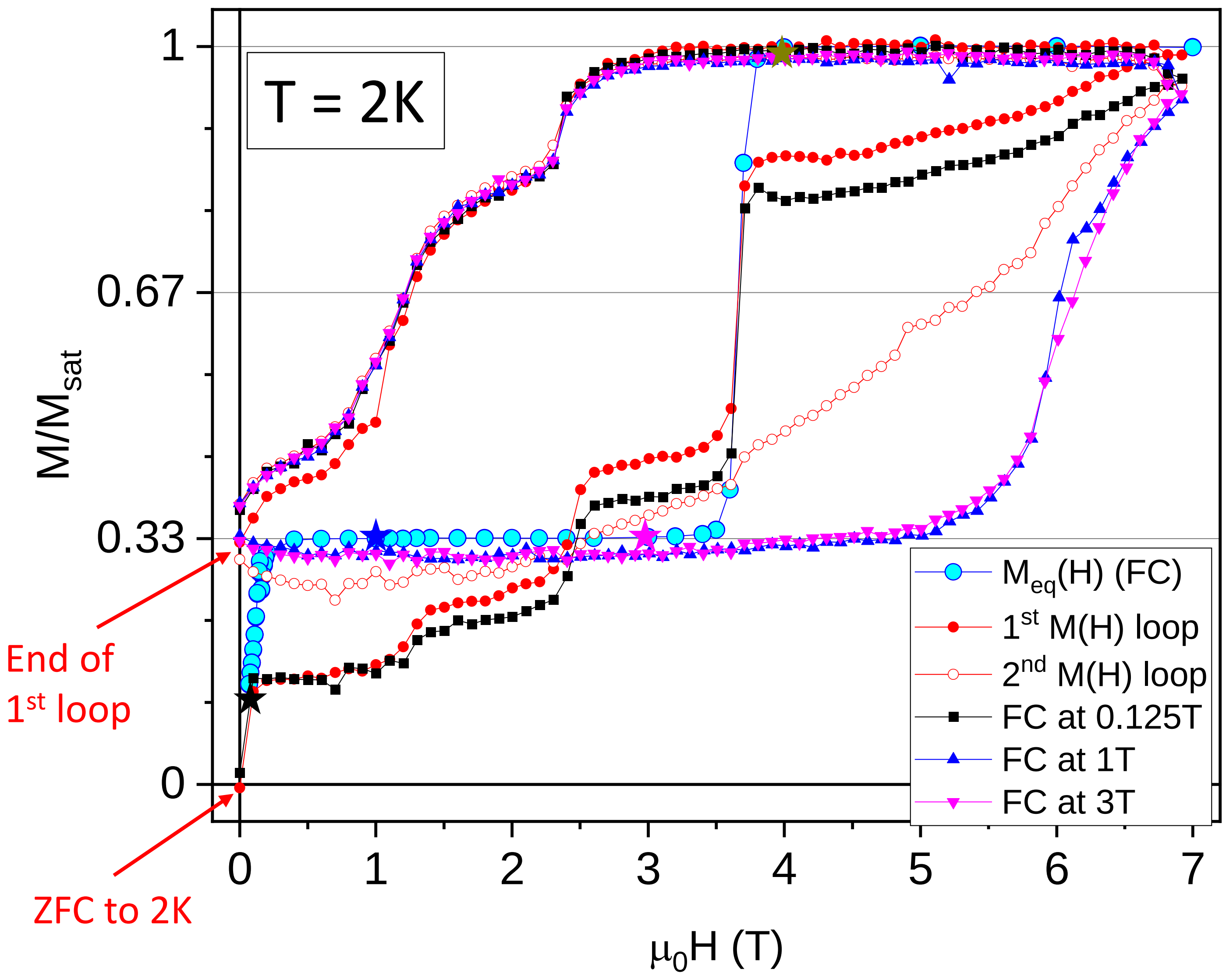
- FC: ordered ground state
- First M(H) loop after ZFC: steps
- Subsequent loops: no steps
- FC at 0.125T → H=0 → loop: similar to 1st loop
- FC at 1T → H=0 → loop:

Magnetic history dependence: How do we access the different states?



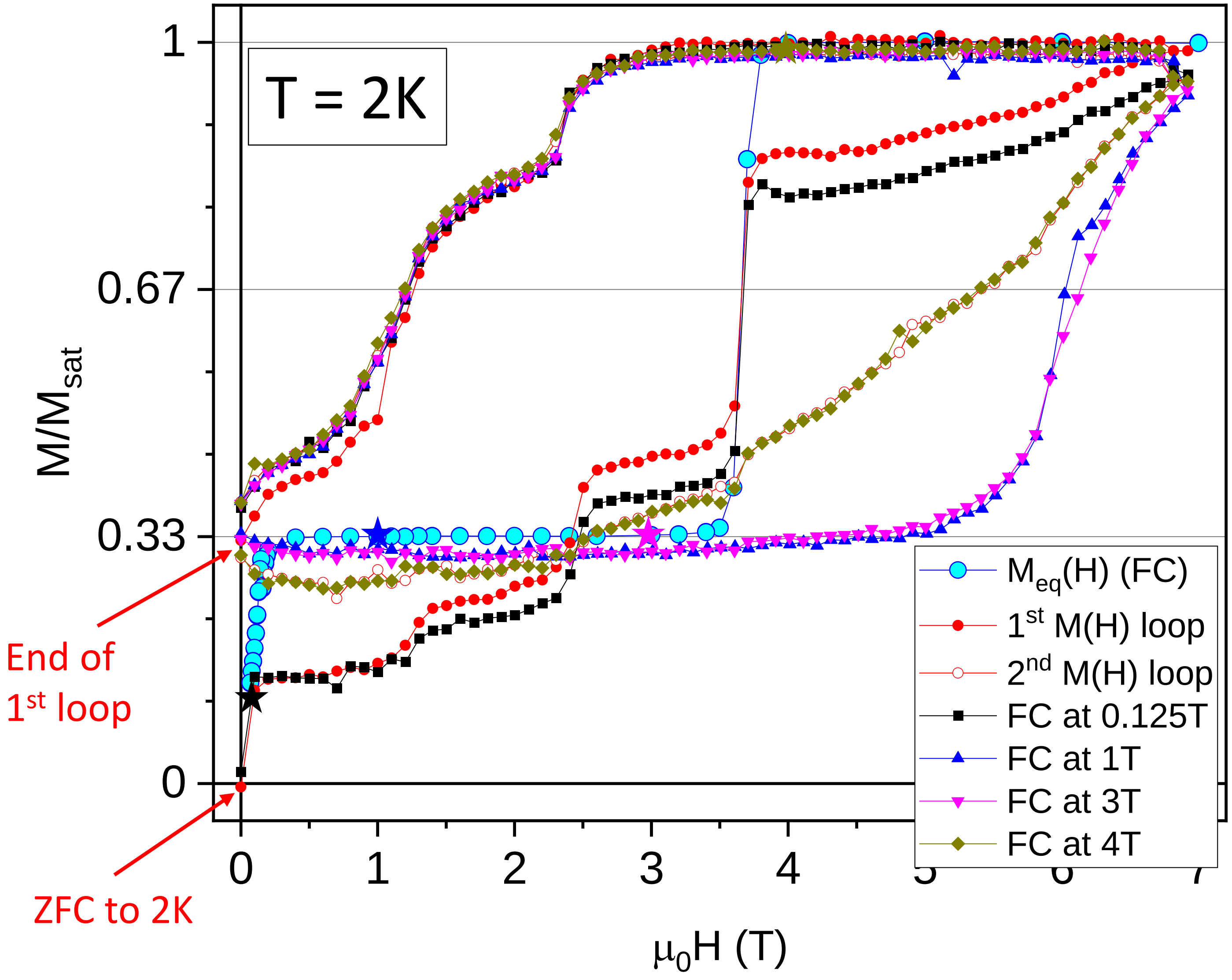
- FC: ordered ground state
- First M(H) loop after ZFC: steps
- Subsequent loops: no steps
- FC at 0.125T → H=0 → loop: similar to 1st loop
- FC at 1T → H=0 → loop: lower branch ~ $M_{sat}/3$
- FC at 3T → H=0 → loop:

Magnetic history dependence: How do we access the different states?



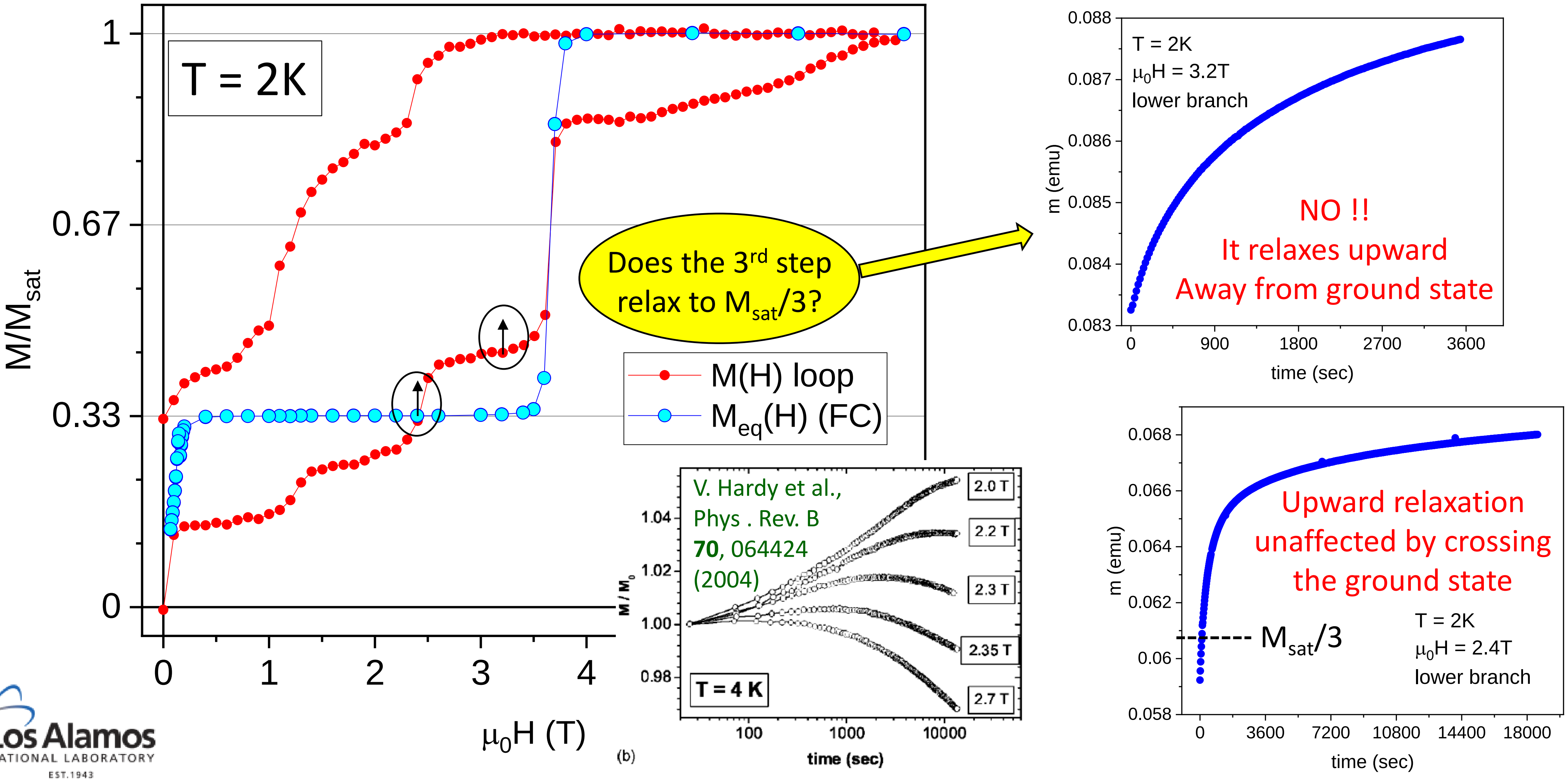
- FC: ordered ground state
- First M(H) loop after ZFC: steps
- Subsequent loops: no steps
- FC at 0.125T → H=0 → loop: similar to 1st loop
- FC at 1T → H=0 → loop: lower branch ~ $M_{sat}/3$
- FC at 3T → H=0 → loop: lower branch ~ $M_{sat}/3$
- FC at 4T → H=0 → loop:

Magnetic history dependence: How do we access the different states?

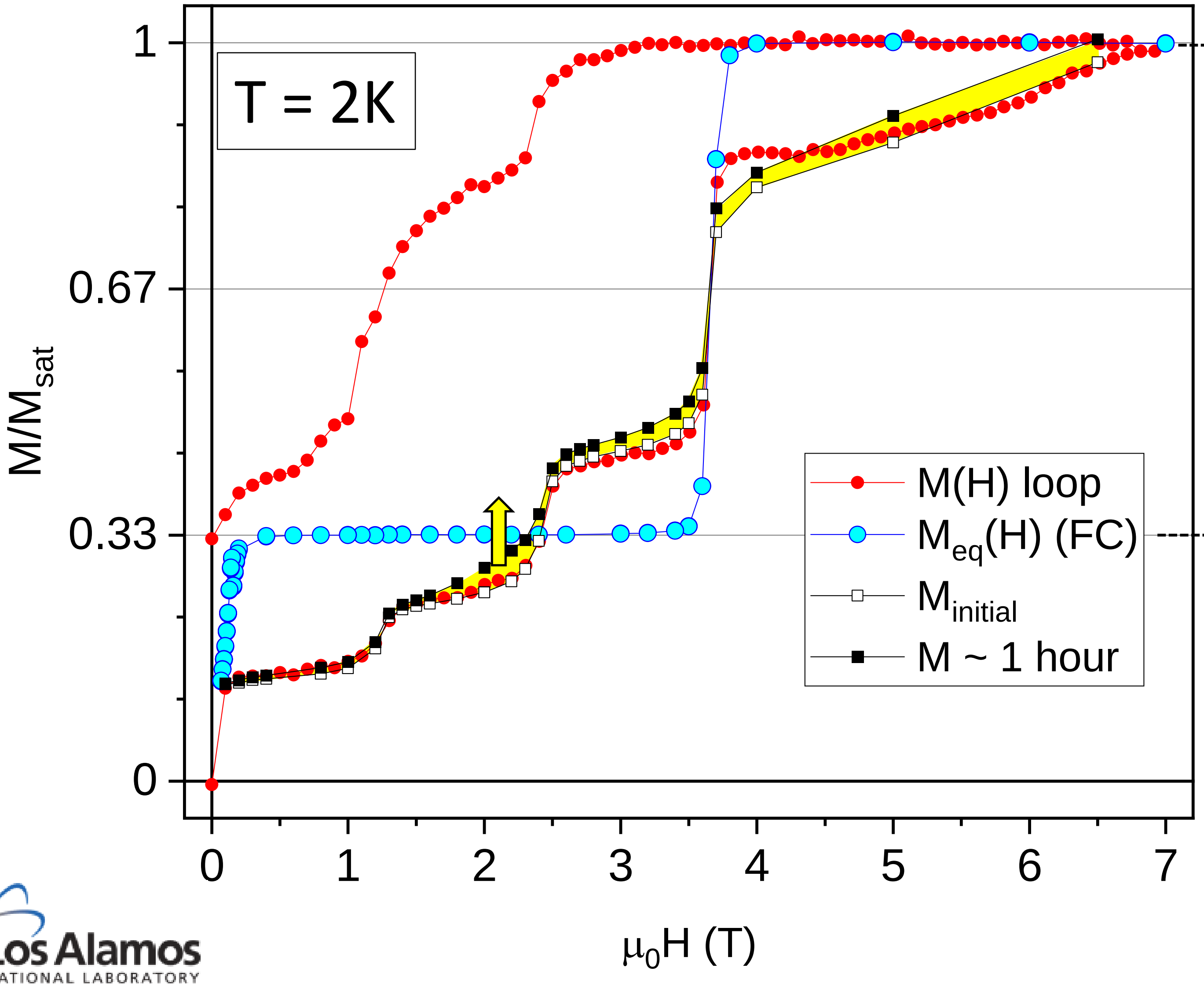


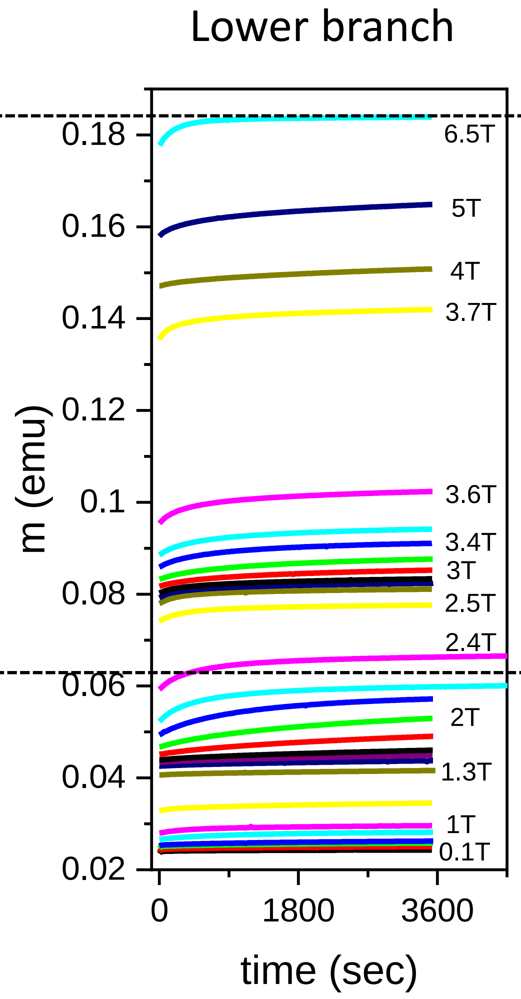
- FC: ordered ground state
- First M(H) loop after ZFC: steps
- Subsequent loops: no steps
- FC at 0.125T → H=0 → loop: similar to 1st loop
- FC at 1T → H=0 → loop: lower branch ~ $M_{sat}/3$
- FC at 3T → H=0 → loop: lower branch ~ $M_{sat}/3$
- FC at 4T → H=0 → loop: same as 2nd loop

Focus has been on understanding the steps in the M(H) loops
Metastable states ⇒ complex
Alternative: to what states are the steps relaxing to?

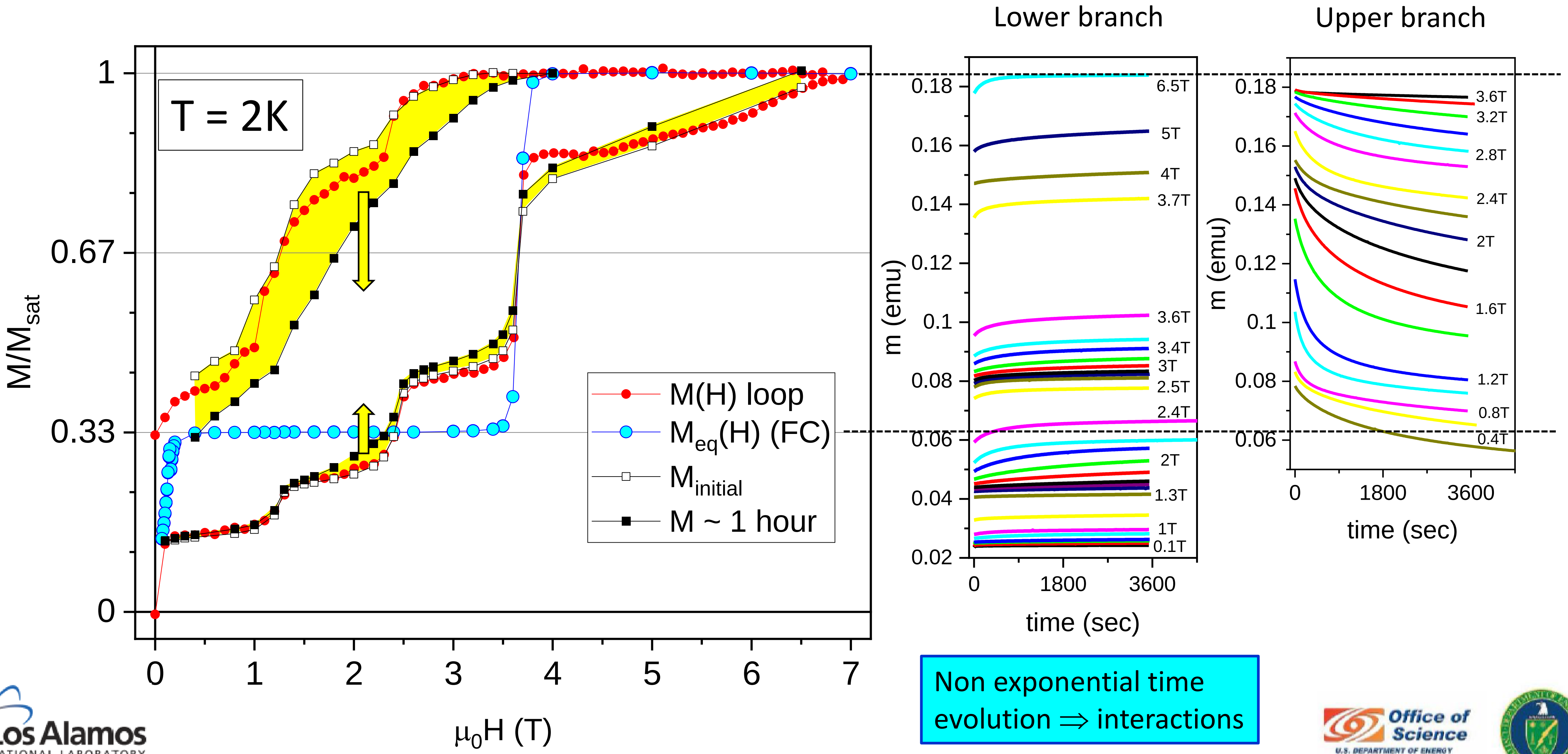


The lower branch of M(H) relaxes upward, for all H (at T = 2K)



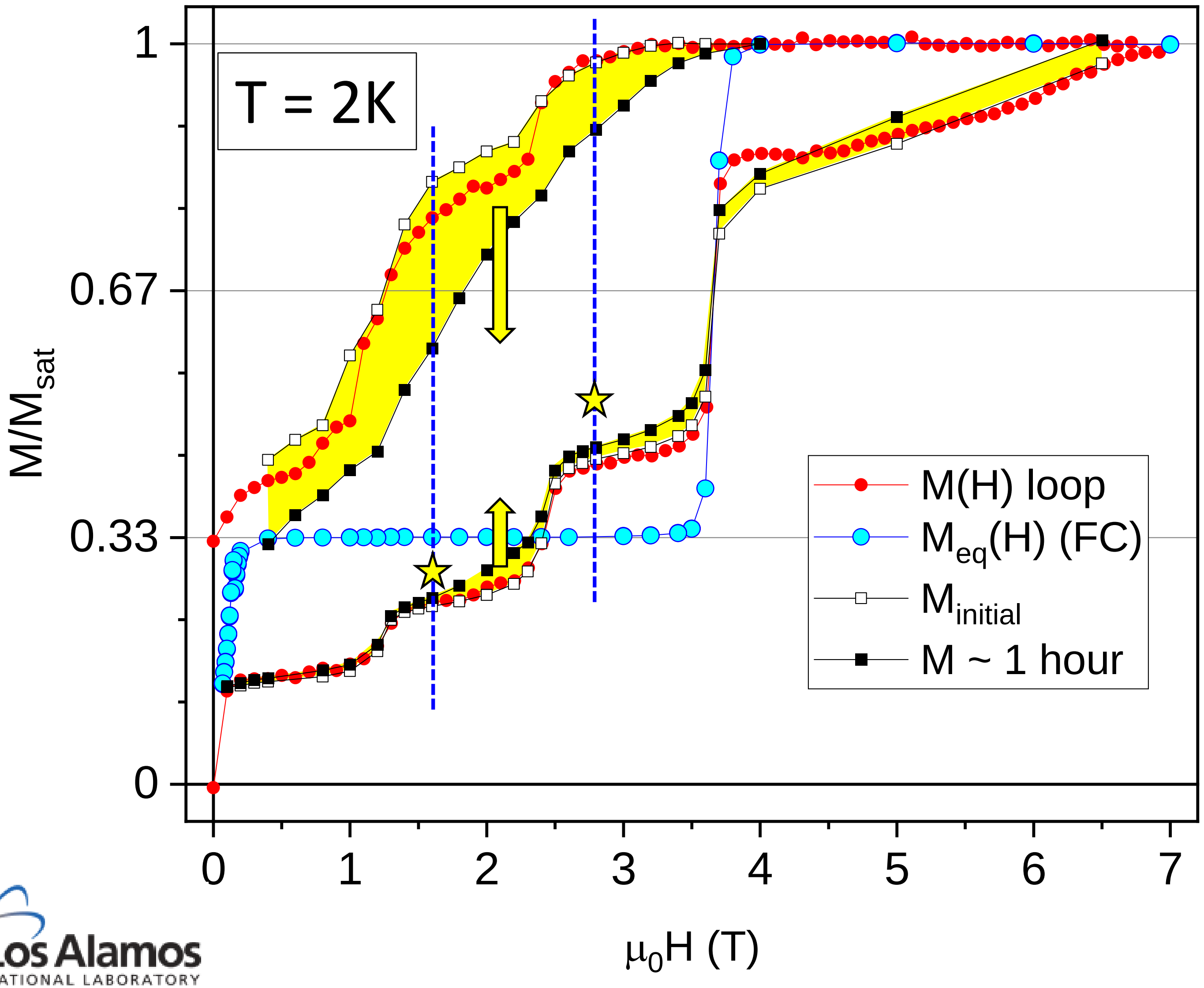


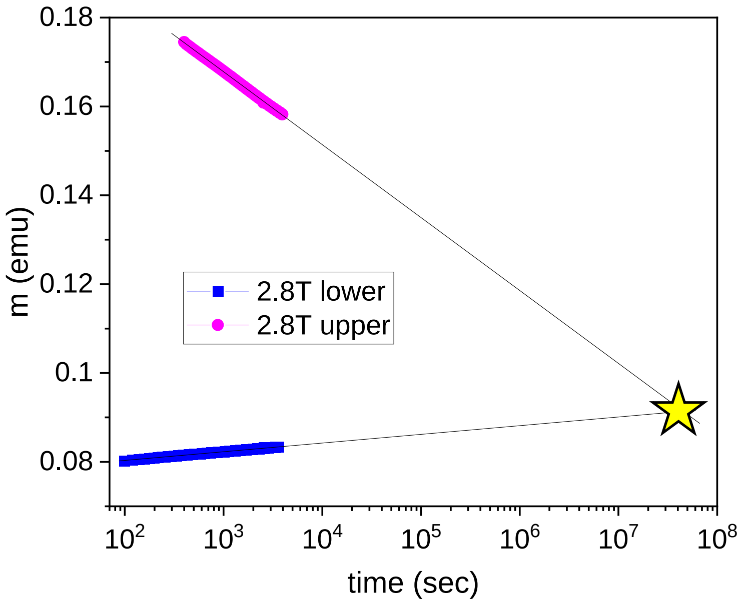
The lower branch of M(H) relaxes upward, for all H (at T = 2K)
The upper branch relaxes downward for all H, and much faster

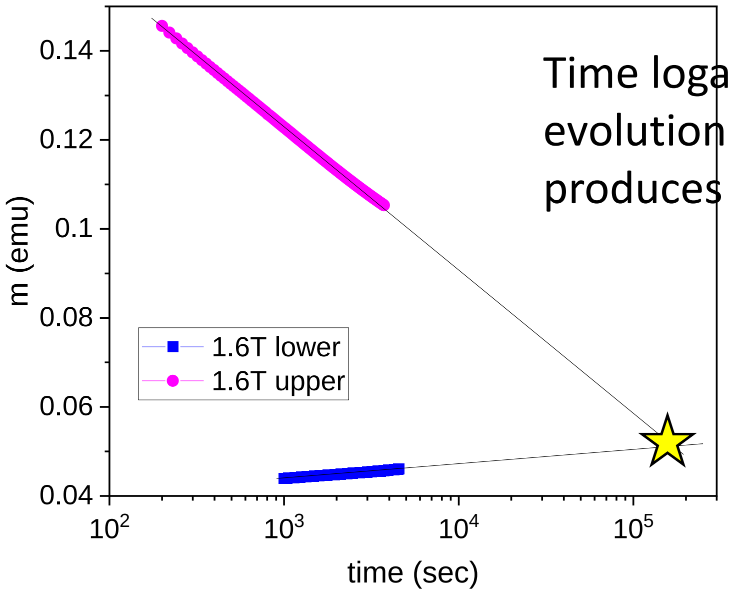


Non exponential time evolution $\Rightarrow$ interactions

Simplest hypothesis:
The lower and upper branches relax to the same state





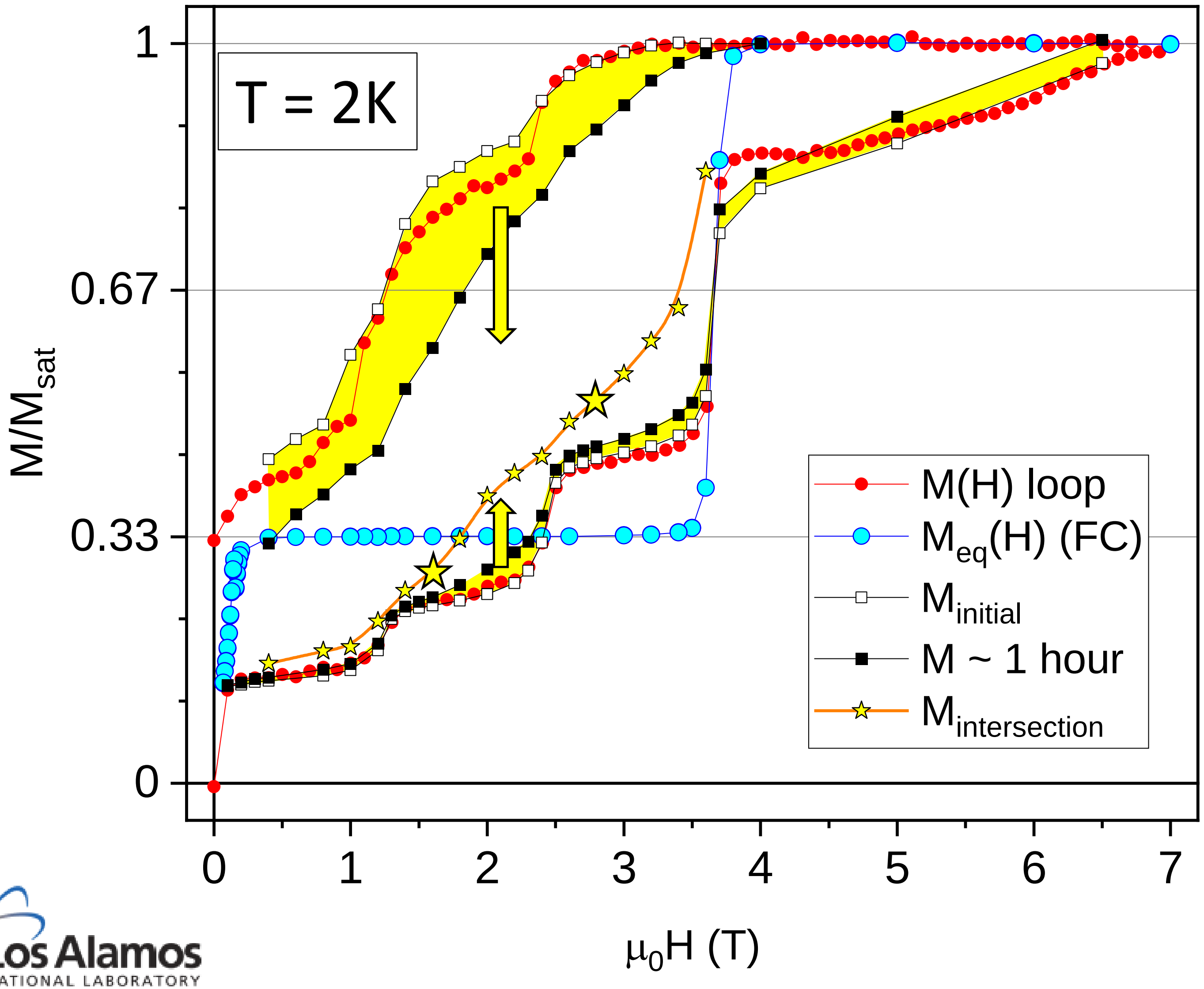


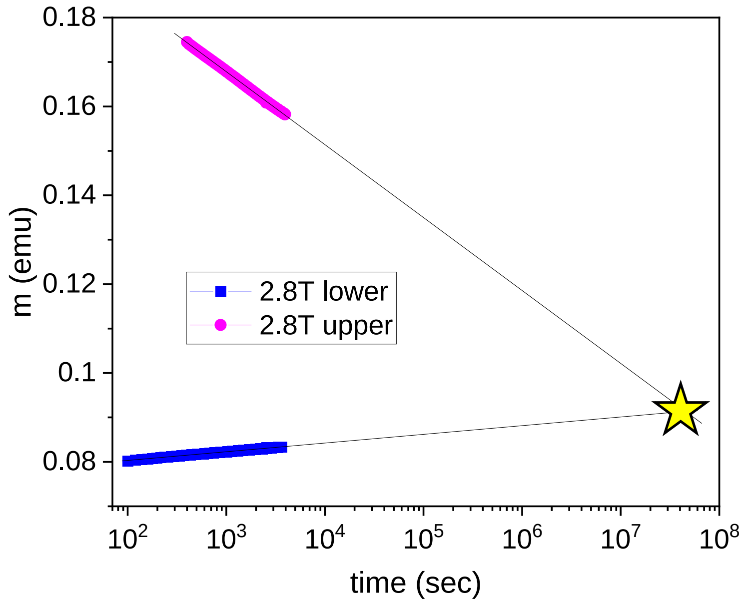
Time logarithmic evolution (vortex-like) produces the best fits!

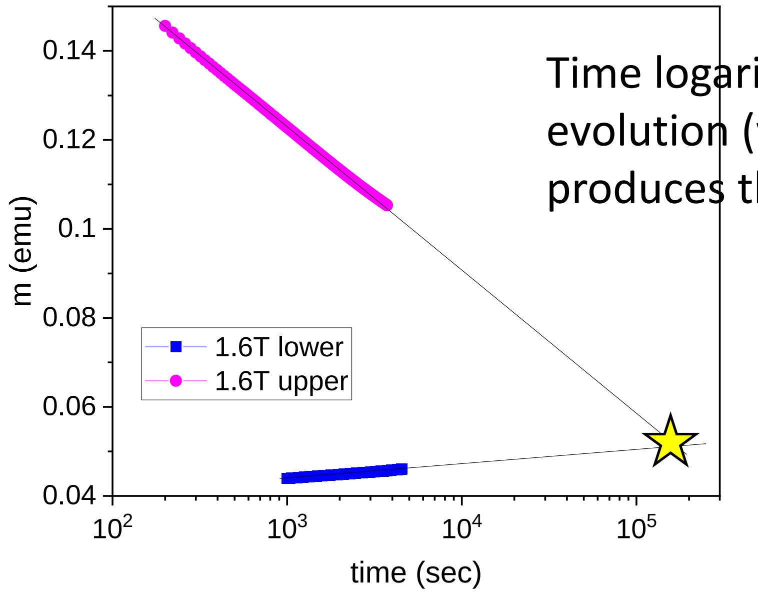
Los Alamos NATIONAL LABORATORY EST.1943

Office of Science U.S. DEPARTMENT OF ENERGY

Evidence suggests that there is a line of lower energy (more stable) states inside the M(H) loop





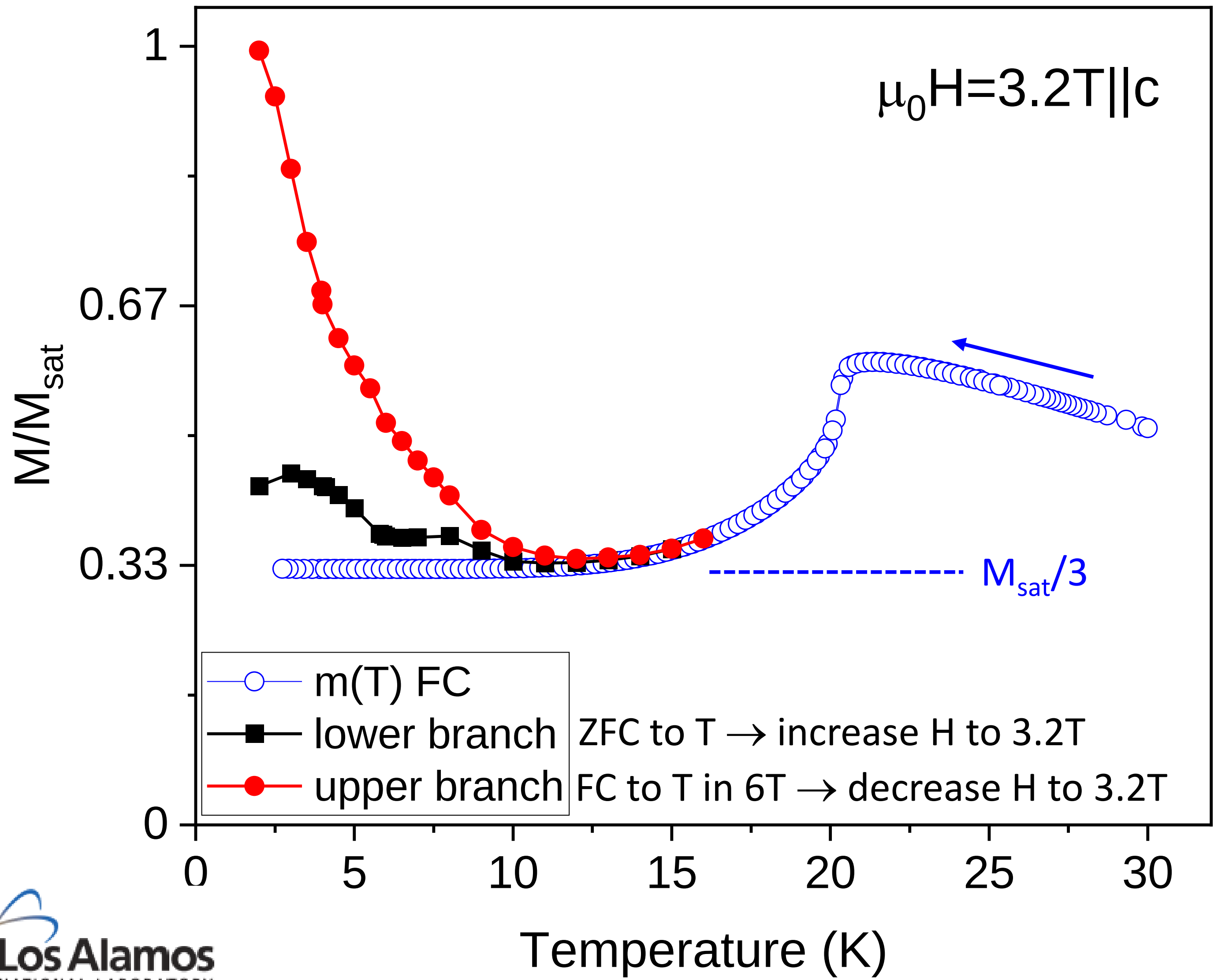


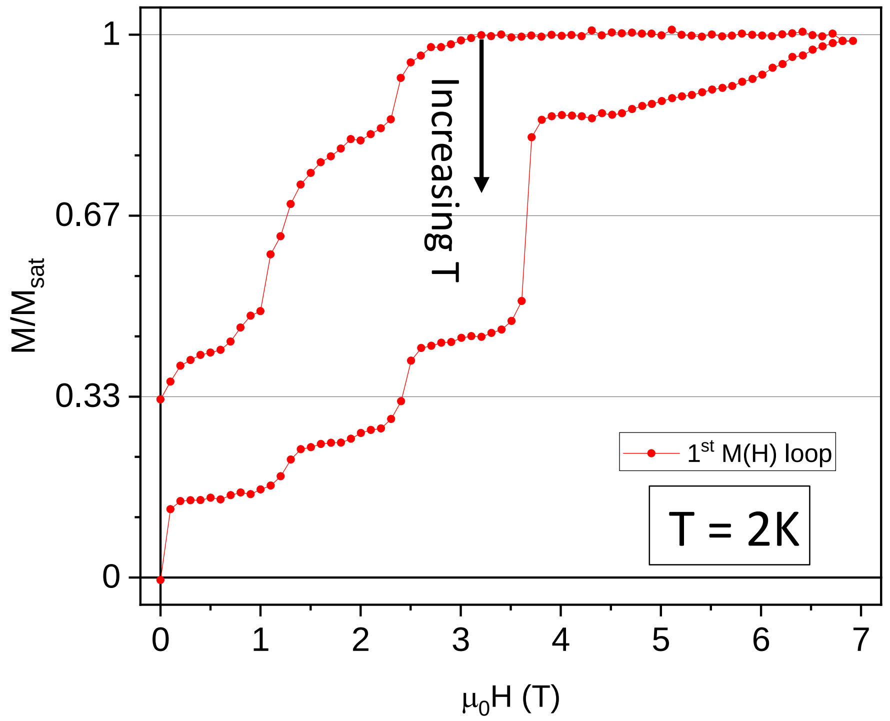
Time logarithmic evolution (vortex-like) produces the best fits!

Can we access intermediate states between both branches, and show that they are more stable?

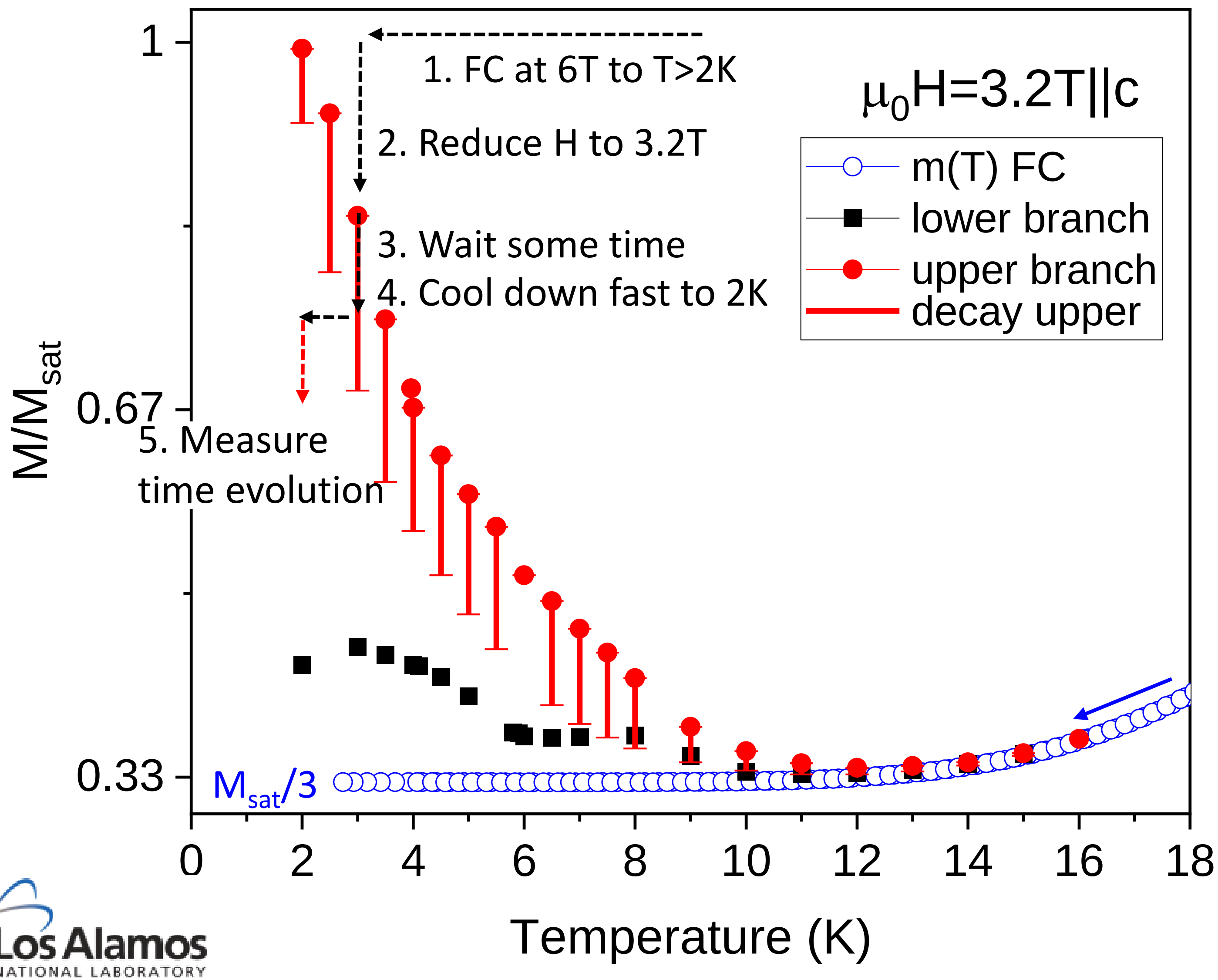
Los Alamos NATIONAL LABORATORY EST.1943

Manipulating the upper branch magnetization by increasing temperature

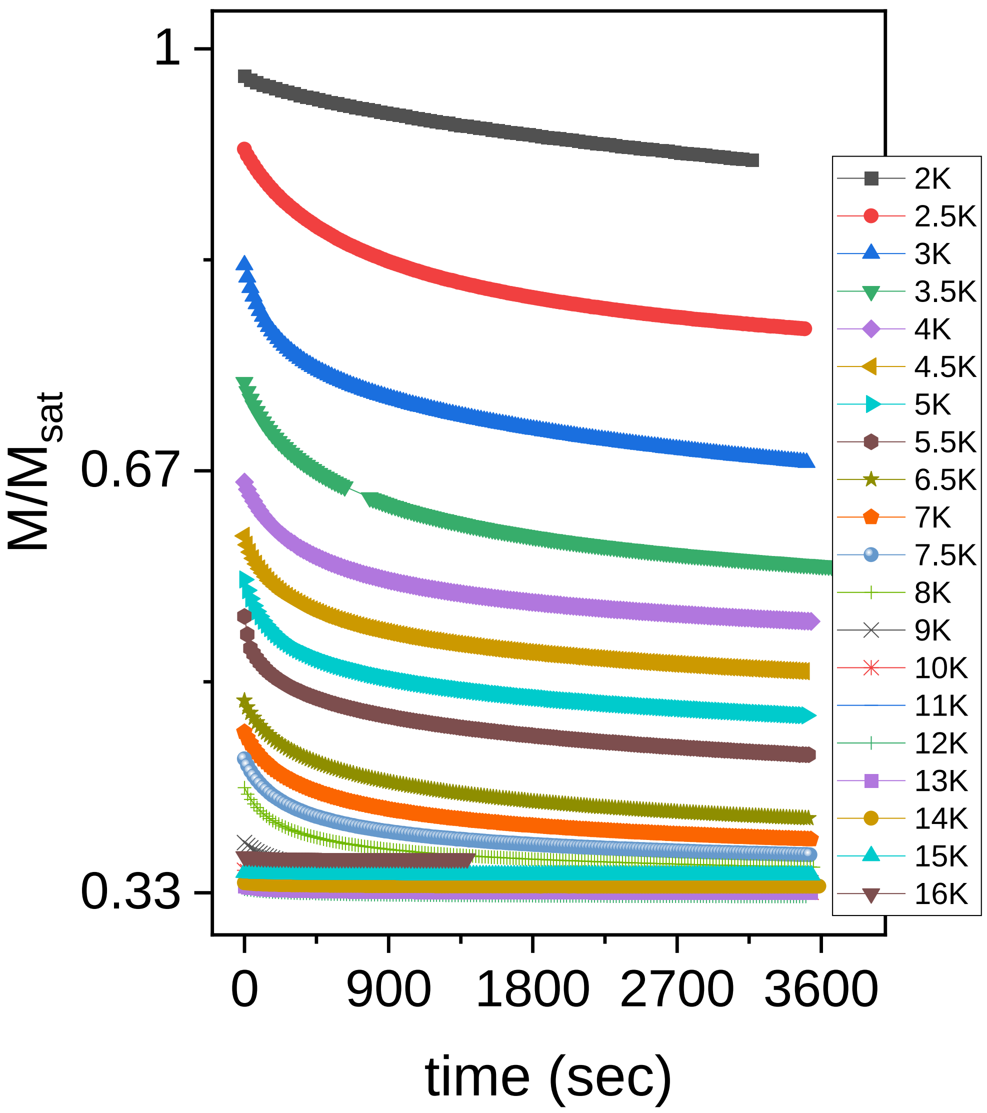




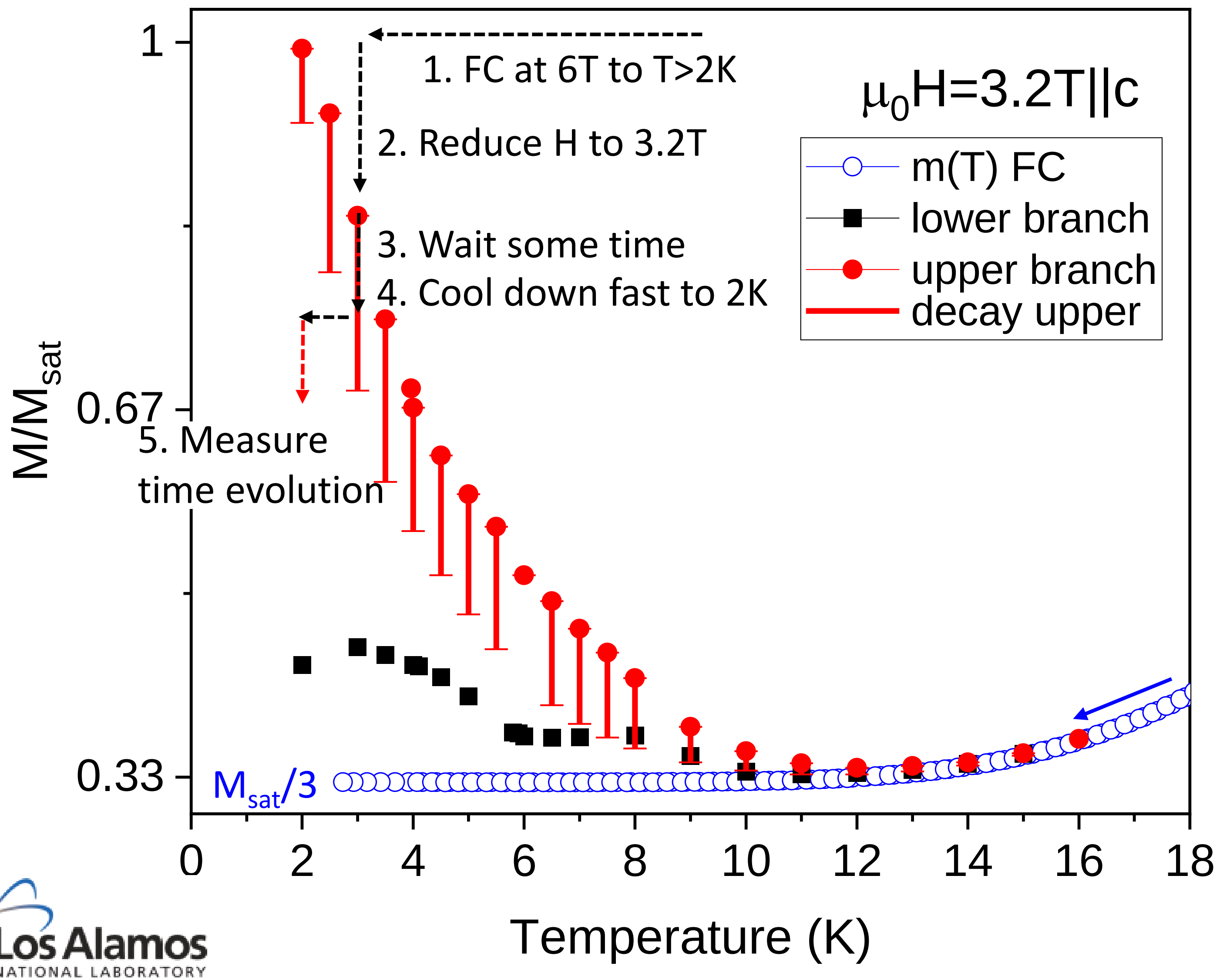
Manipulating the upper branch magnetization by increasing temperature



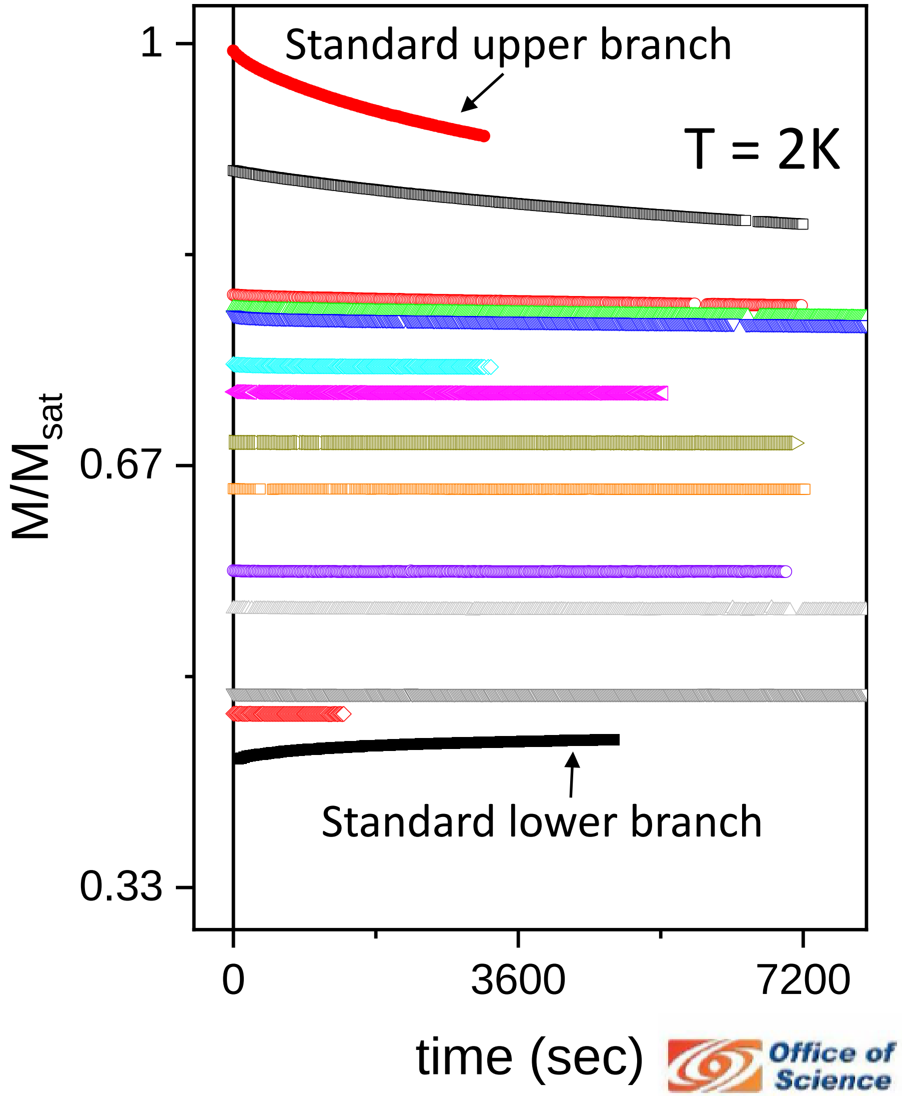
Time dependence upper branch: fast downward evolution at all T



Manipulating the upper branch magnetization by increasing temperature



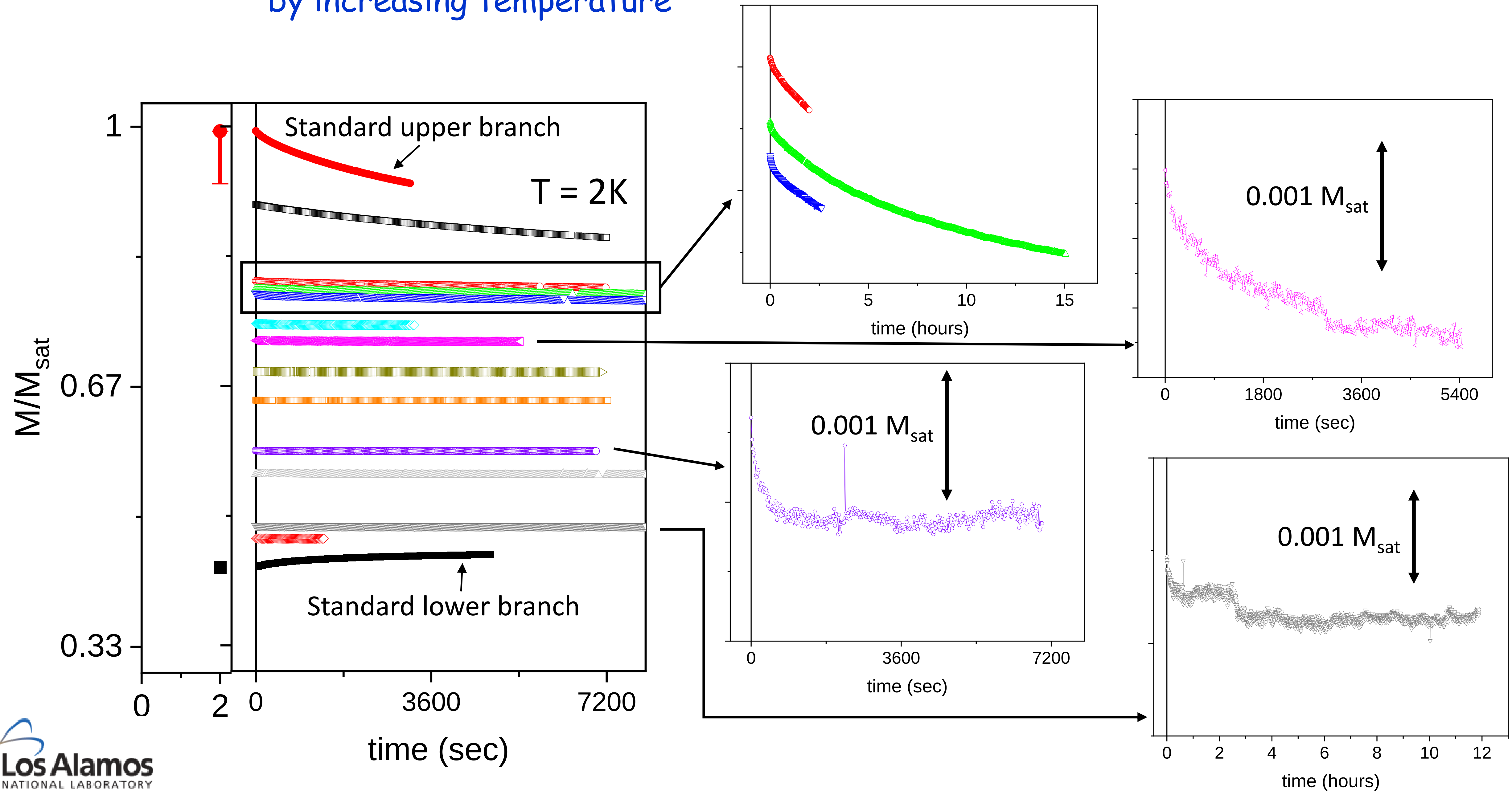
Time dependence upper branch at 2K, with initial M set by magnetic history



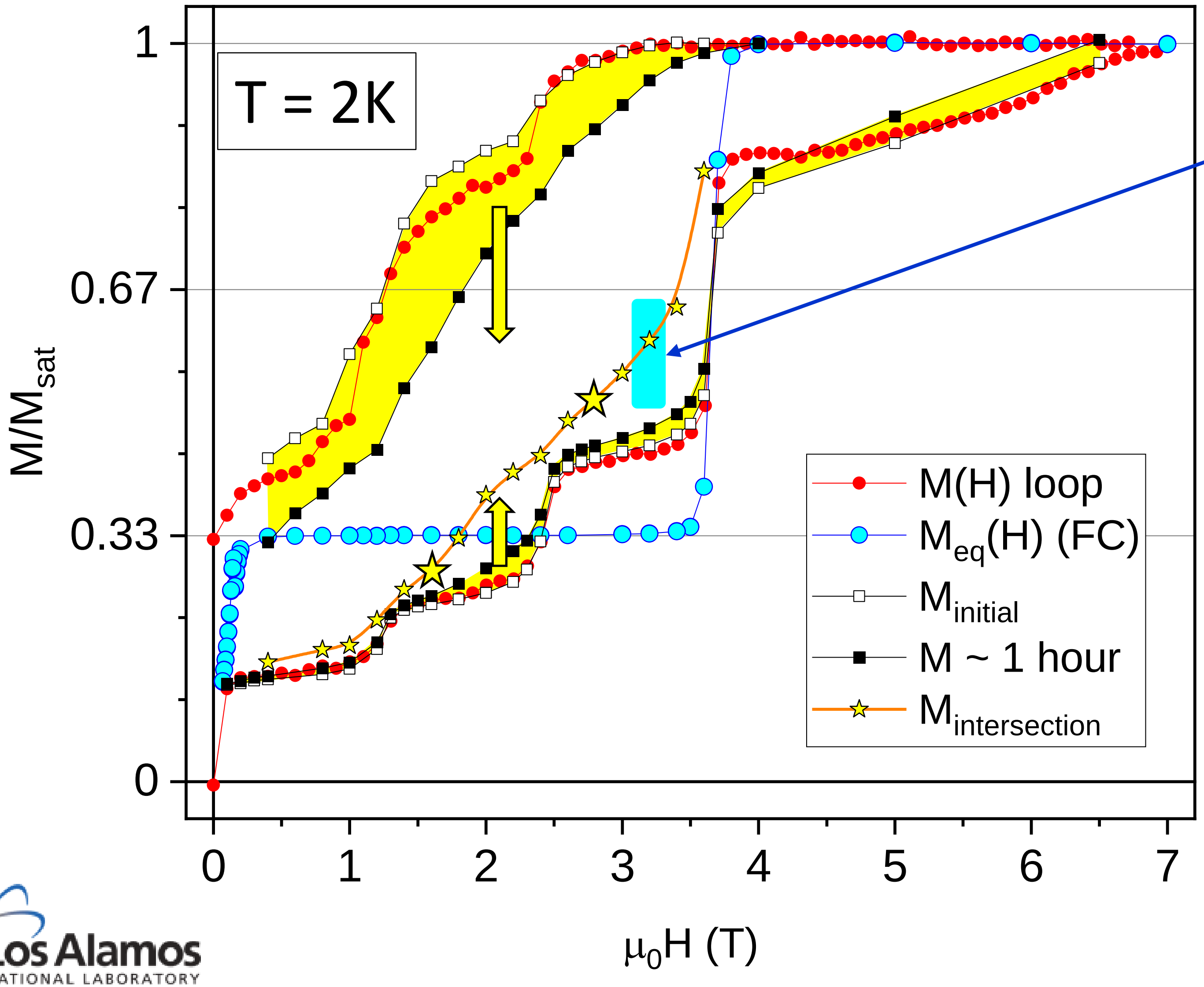
Los Alamos NATIONAL LABORATORY EST.1943

Office of Science U.S. DEPARTMENT OF ENERGY

Manipulating the upper branch magnetization by increasing temperature



There is a region inside the M(H) loop where the magnetization is more stable than in the lower and upper branches



At T = 2K and $\mu_0 H$ = 3.2 T, there are states in a range of M intermediate between both branches that are stable within our experimental resolution

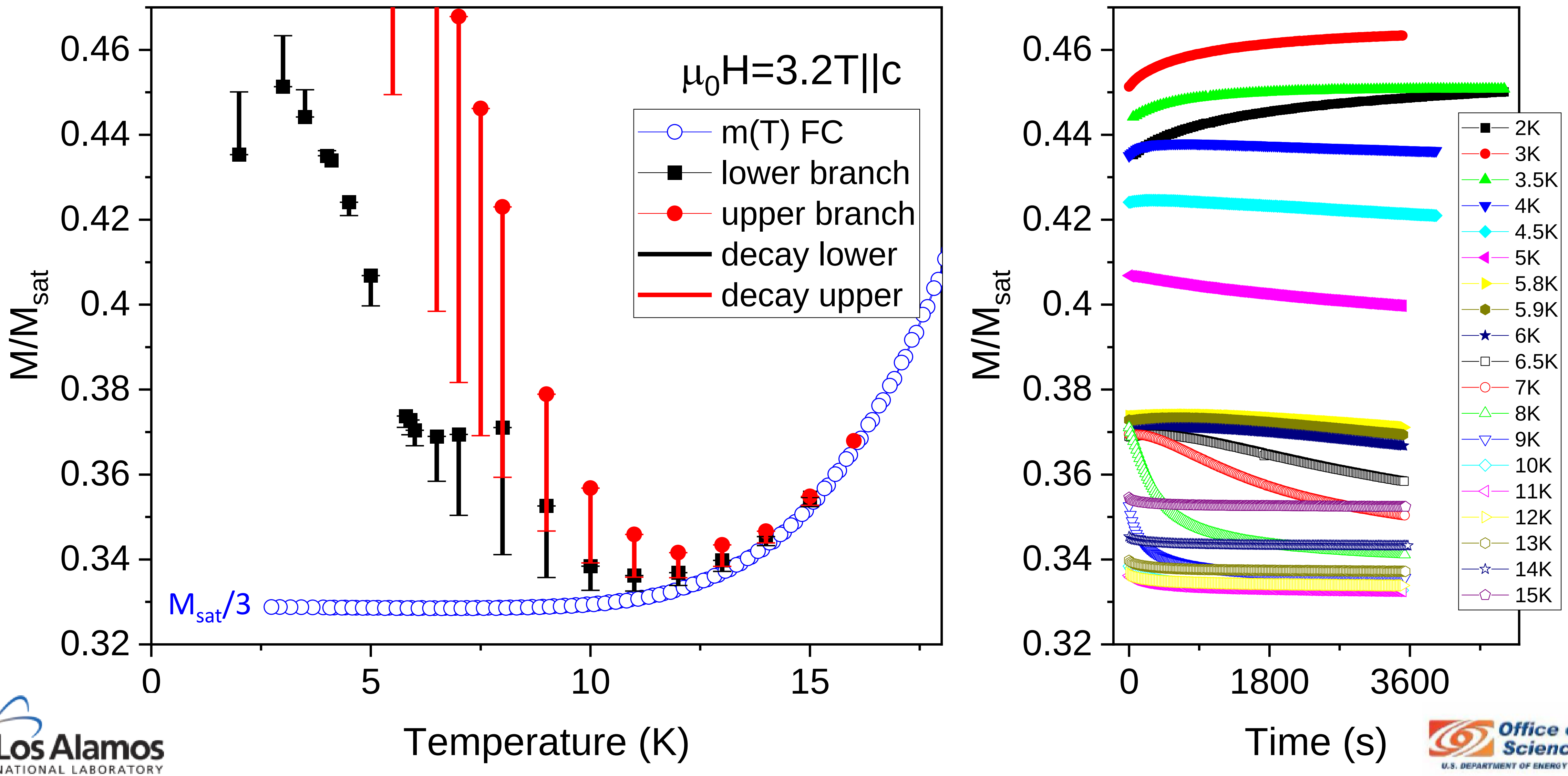
Work in progress: explore the location of the stability region at other fields

Does this stability region extend to higher temperatures?

Los Alamos NATIONAL LABORATORY EST.1943

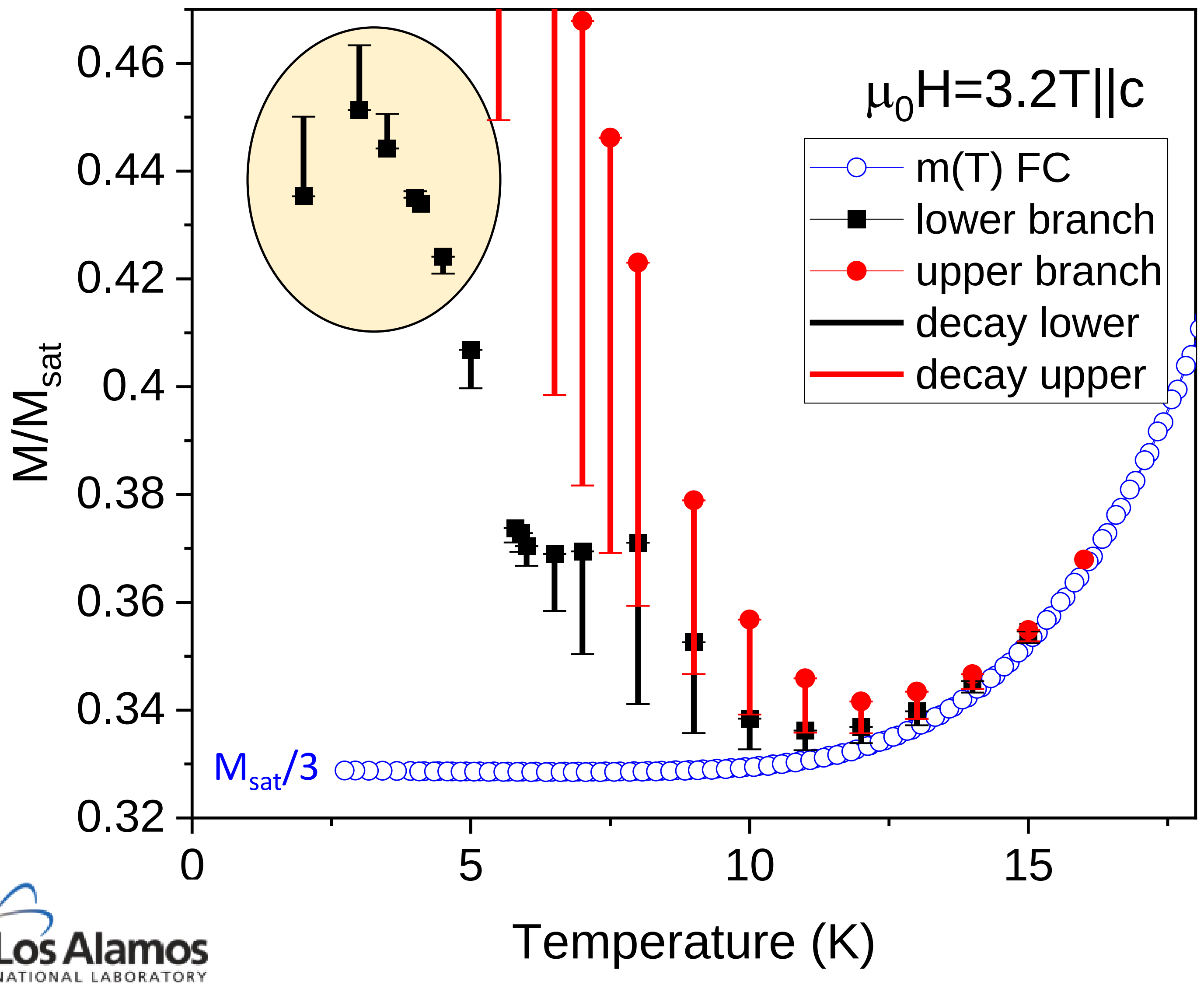
Office of Science U.S. DEPARTMENT OF ENERGY

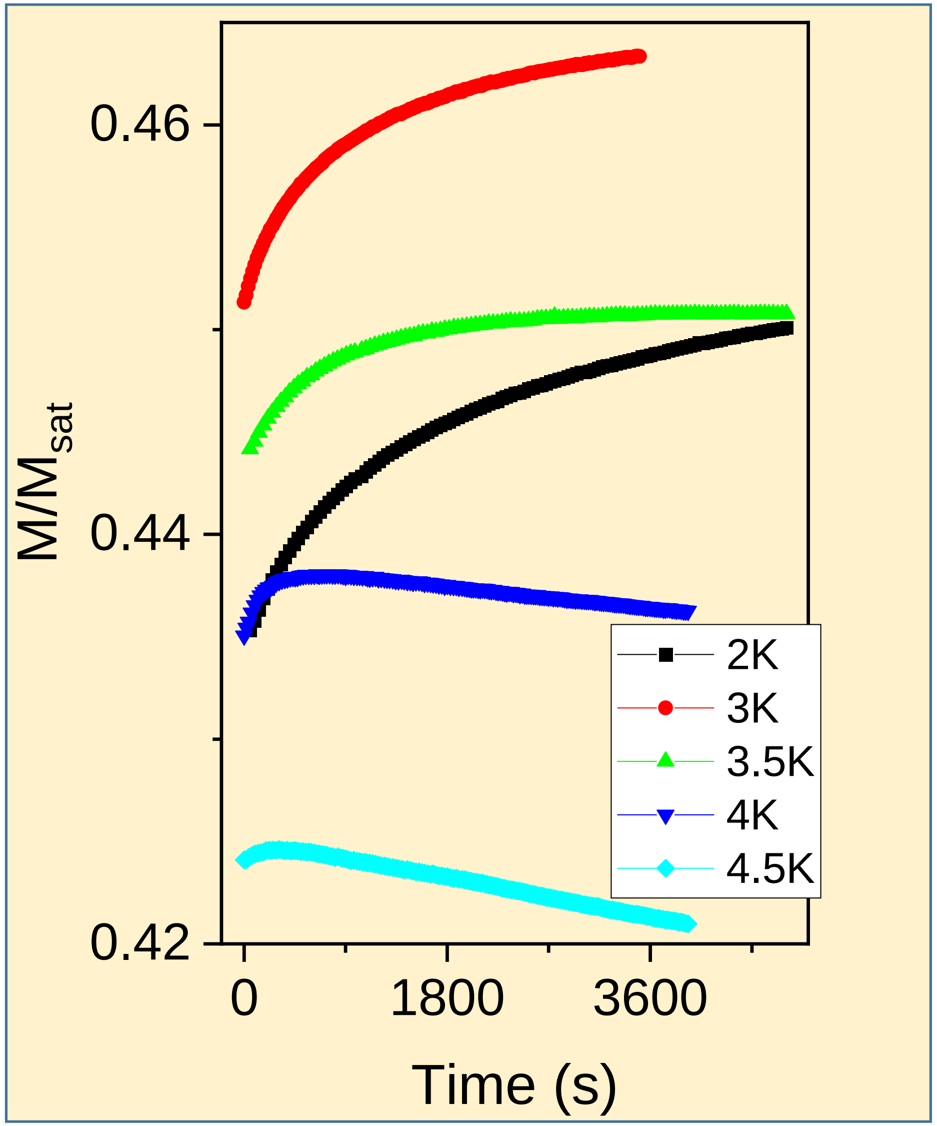
The temperature dependence of the time evolution of the lower branch is quite complex



The temperature dependence of the time evolution of the lower branch is quite complex

The upward relaxation remains at T higher than 2 K, but around 3.5 K the evolution becomes non-monotonic (in our timescales) ⇒ the central stability region dissapears

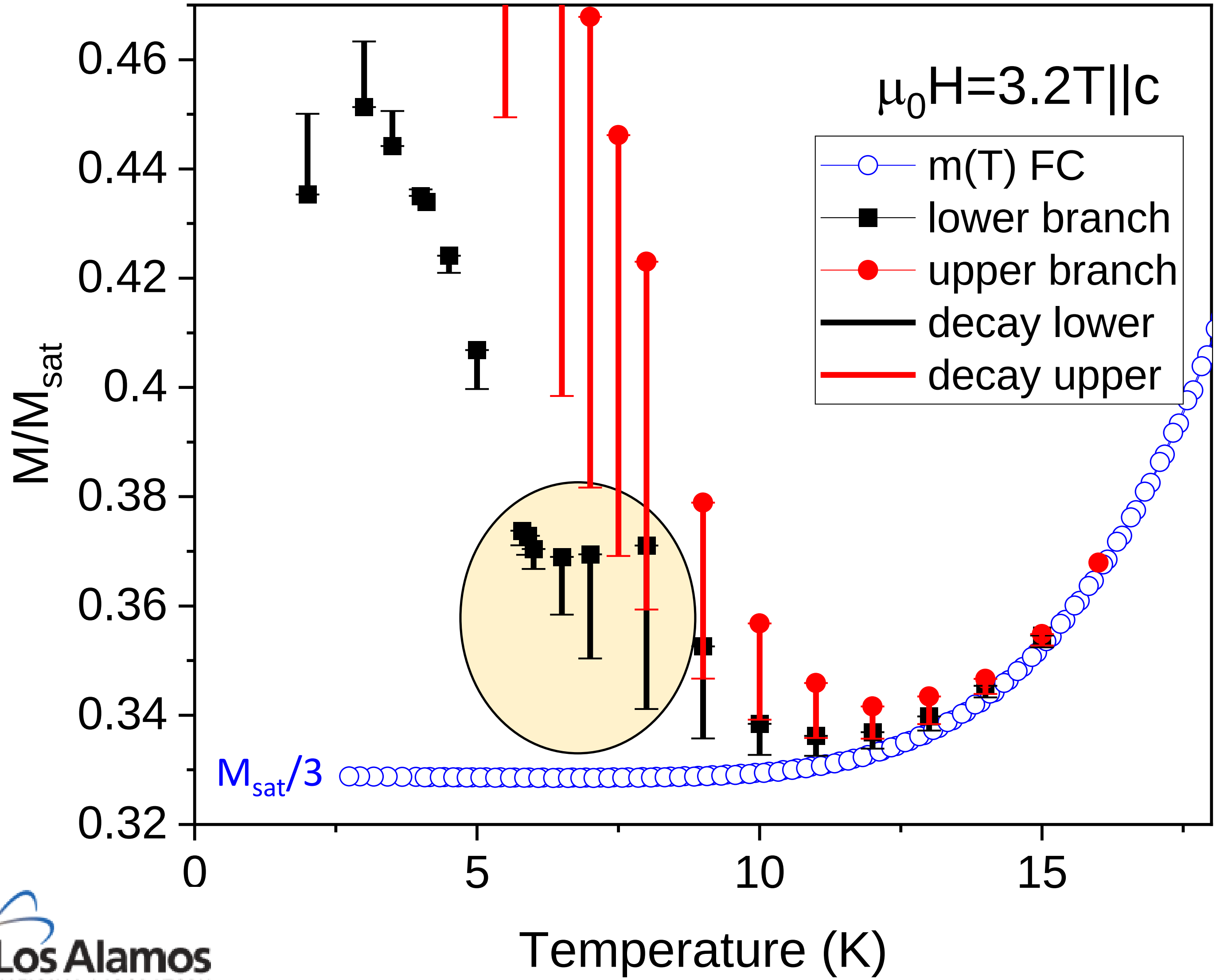


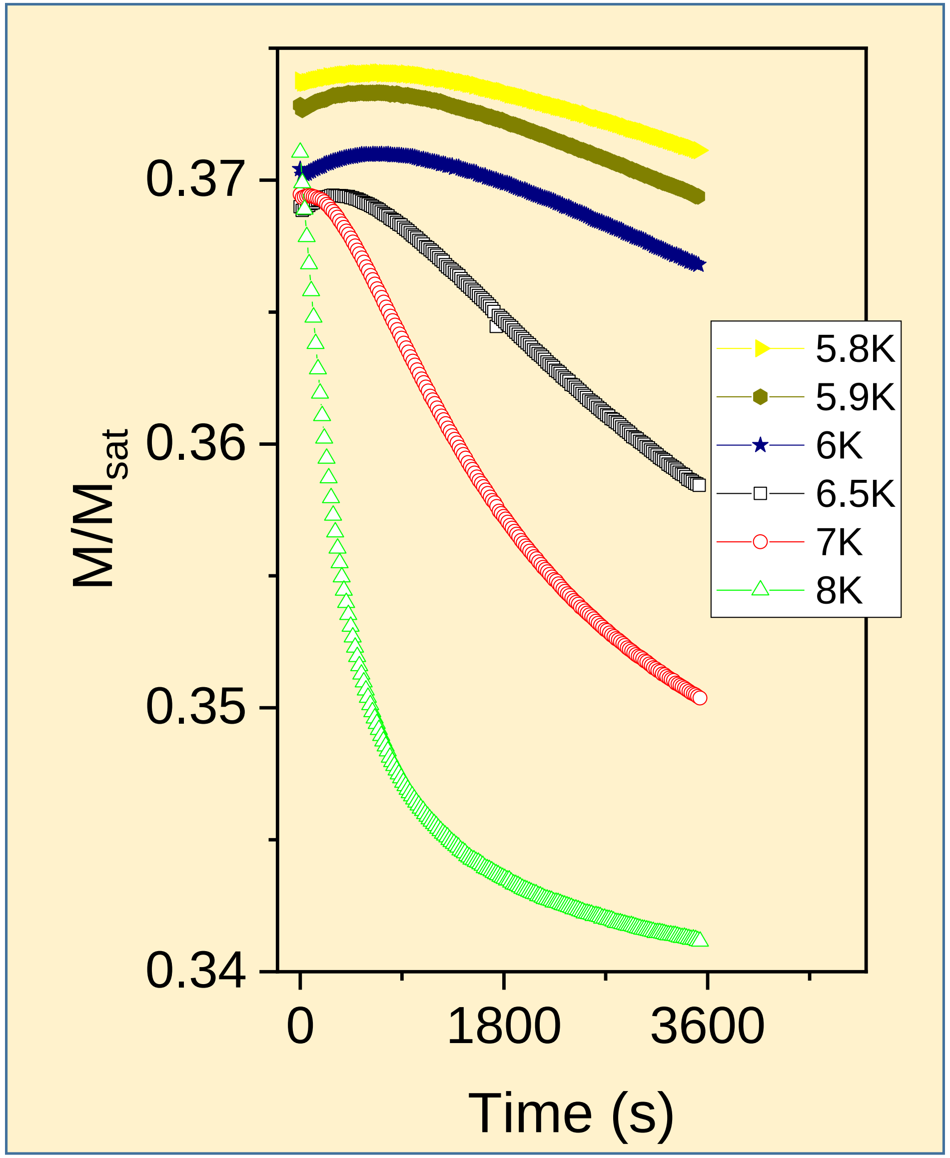


Los Alamos
NATIONAL LABORATORY
EST.1943

The temperature dependence of the time evolution of the lower branch is quite complex

The non-monotonic evolution reappears in the 5 K to 7 K range.

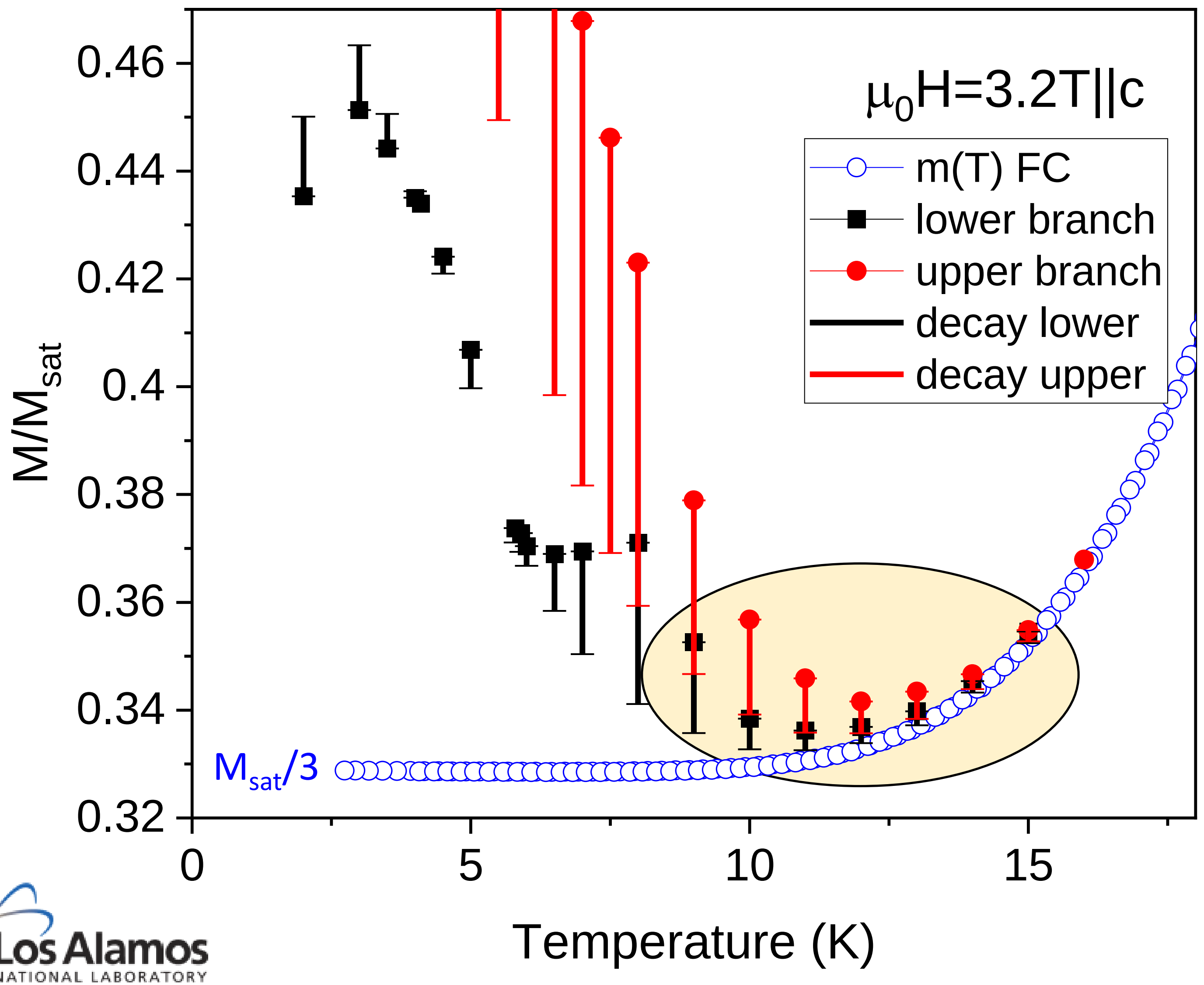


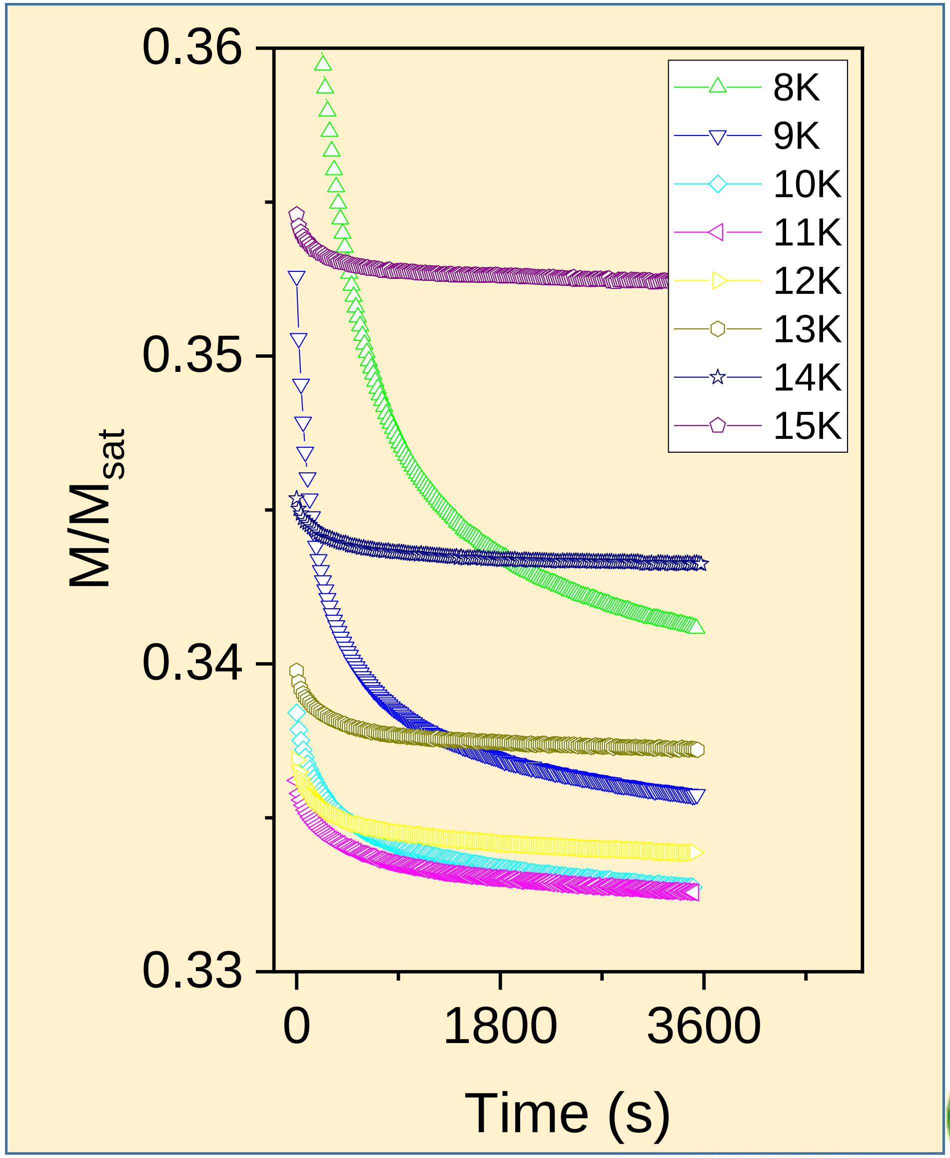


Los Alamos
NATIONAL LABORATORY
EST. 1943

The temperature dependence of the time evolution of the lower branch is quite complex

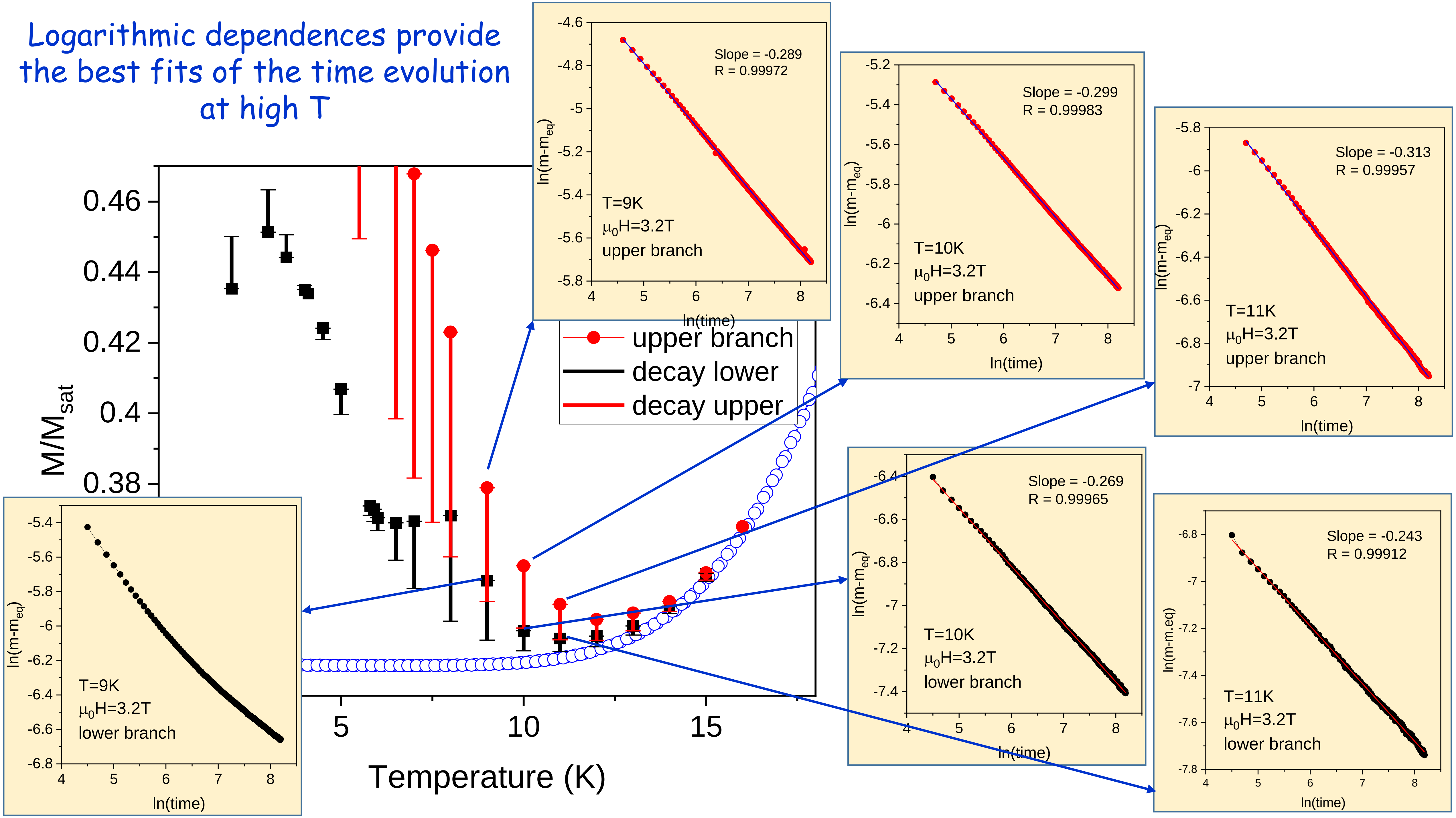
Above ~ 9 K, a simpler picture emerges:
monotonic downward time evolution towards the ground state, no evidence of other phases



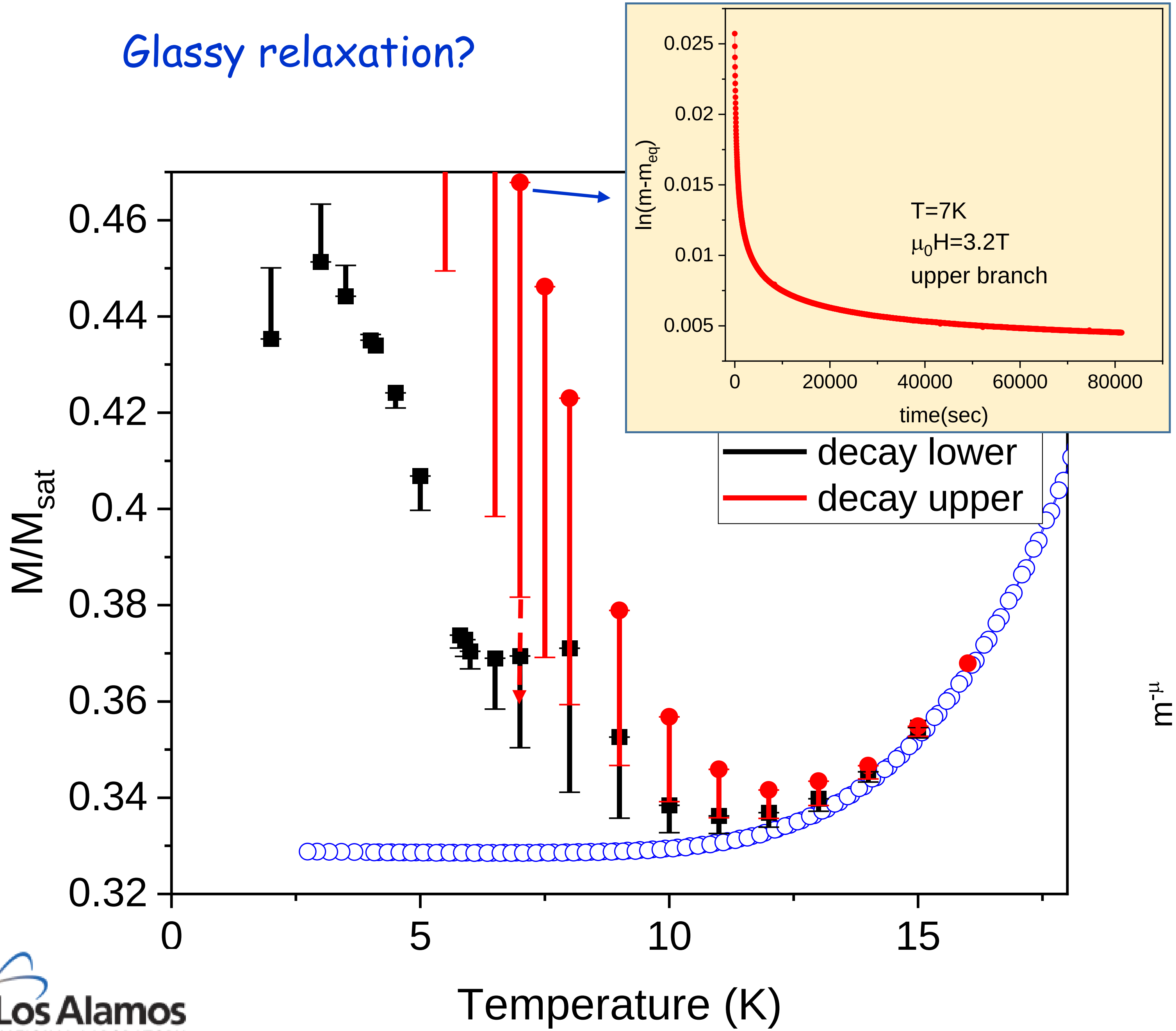


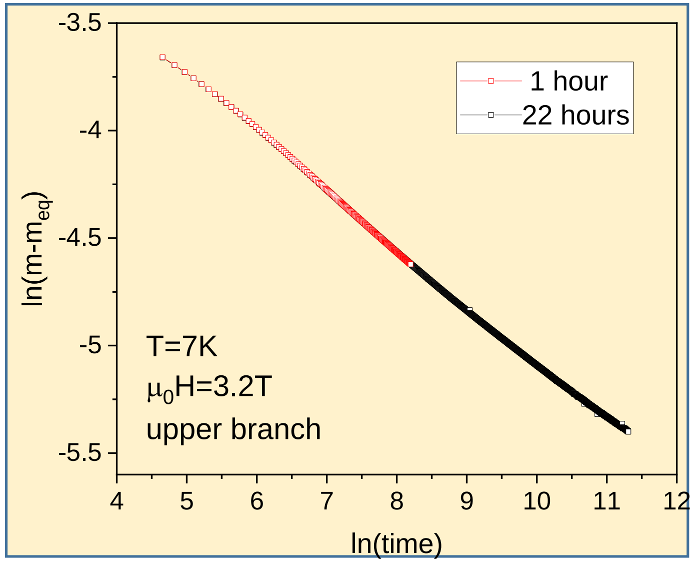
Los Alamos NATIONAL LABORATORY EST.1943

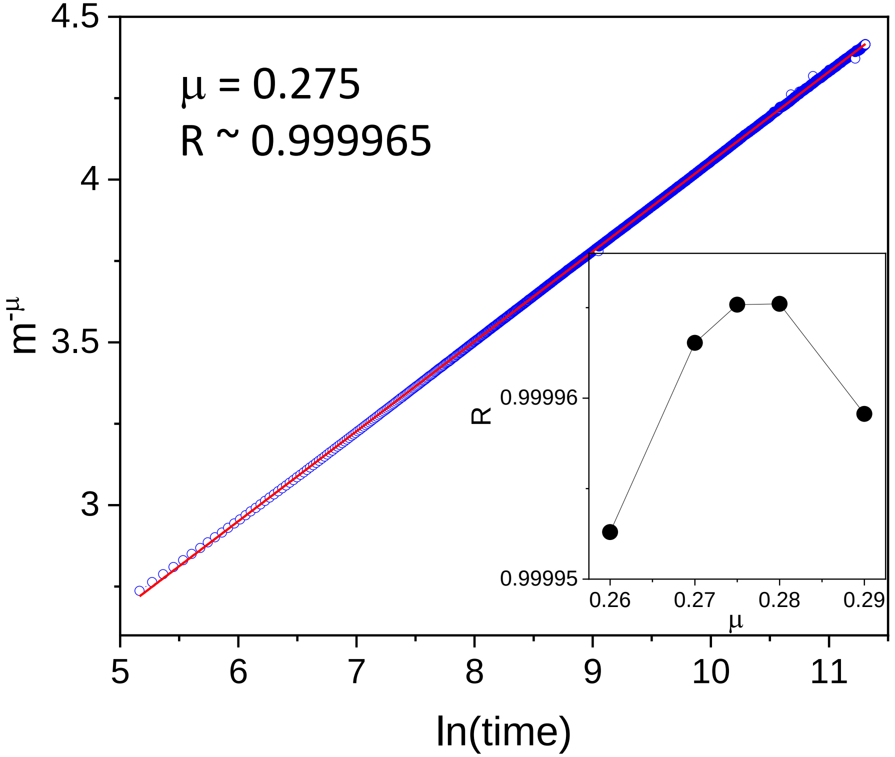
Logarithmic dependences provide the best fits of the time evolution at high T



Glassy relaxation?







Los Alamos
NATIONAL LABORATORY
EST.1943

Summary

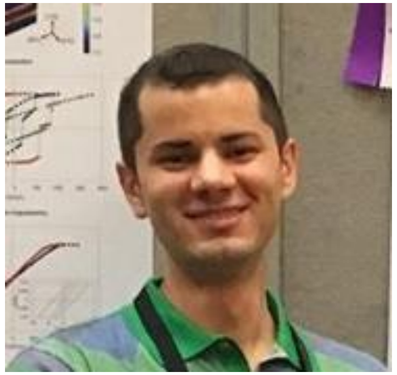
Over the last several decades, we at the superconductivity research community have developed a powerful set of experimental and theoretical tools to investigate vortex matter.

Applying those tools to other materials and phenomena, as in the example presented in this talk, can provide a fresh perspective to solve some problems.

The magnetic properties of the geometrically frustrated spin-chain compound $Ca_3Co_2O_6$ are complex, and a complete description of the steps in the M(H) loops remains elusive. We have identified a previously unobserved stable phase at low T. A simpler vortex-like dynamics emerges at high T.

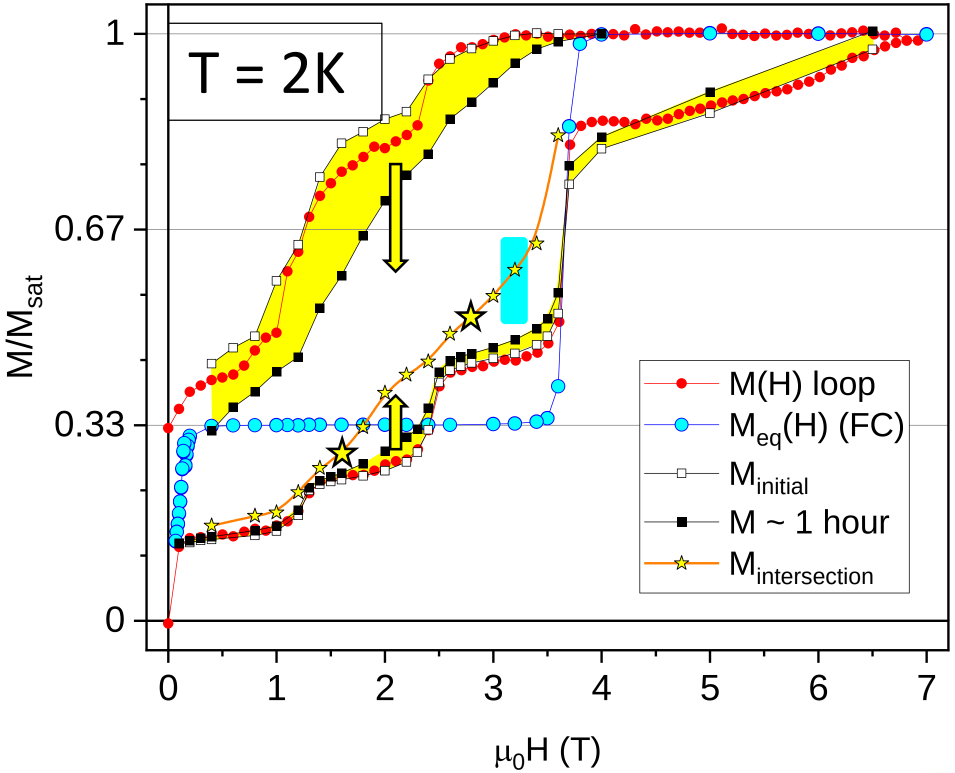
Thank you to the organizers for working so hard to put this workshop together, in spite of all the challenges!!

Looking forward to see all of you (I mean, <u>really</u> see you) in the next workshop!



Ivan Nekrashevich
LANL

Vivien Zapf
LANL



Los Alamos NATIONAL LABORATORY EST.1943

Office of Science U.S. DEPARTMENT OF ENERGY